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## 1 Assessment of a flamelet approach to evaluating mean species mass fractions in moderately and 2 highly turbulent premixed flames

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### 10 Abstract

11 Complex-chemistry Direct Numerical Simulation (DNS) data obtained from lean methane-air turbulent flames are analysed to  
12 perform *a priori* assessment of predictive capabilities of the flamelet approach to evaluating mean concentrations of various  
13 species in turbulent flames characterized by Karlovitz numbers  $Ka = 6.0$ ,  $74.0$ , and  $540$ . Six definitions of a combustion  
14 progress variable  $c$  are probed and two types of Probability Density Functions (PDFs) are adapted: (i) actual PDFs extracted  
15 directly from the DNS data or (ii) presumed  $\beta$ -function PDFs obtained using the DNS data on the first two moments of the  $c$ -  
16 field. Results show that the mean density, the mean temperature, and the mean mass fractions of  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  
17  $\text{CH}_2\text{O}$ ,  $\text{CH}_3$ , and  $\text{HCO}$  are very well predicted using the temperature-based combustion progress variable  $c_T$  and the actual  
18 PDF. For other considered species, the quantitative predictions are worse, but still appear to be encouraging (with the exception  
19 of  $\text{CH}_3\text{O}$  at  $Ka = 540$ ). The use of the flamelet library obtained from the equidiffusive laminar flame improves results for  $\text{H}_2$ ,  
20  $\text{HO}_2$ , and  $\text{H}_2\text{O}_2$  at the highest Karlovitz number. Alternative definitions of the combustion progress variable perform worse and  
21 the reasons for this are explored. The use of the  $\beta$ -function PDF yields worse results for intermediate species such as  $\text{OH}$ ,  $\text{O}$ ,  
22  $\text{H}$ ,  $\text{CH}_3$ , and  $\text{HCO}$ , with this PDF being significantly different from the actual PDF. Application of the flamelet approach to  
23 rates of production/consumption of various species is also addressed and implications of obtained results for modeling are  
24 discussed.

25 **Keywords:** premixed turbulent combustion, complex chemistry, modeling, DNS, PDF, flamelet

### 26 NOMENCLATURE

27 $a, b$	parameters of beta function PDF
28 $c$	combustion progress variable
29 $Da$	Damköhler number
30 $g = \overline{c'^2} / [\overline{c}(1 - \overline{c})]$	segregation factor
31 $Ka$	Karlovitz number
32 $L_{xx}$	longitudinal integral length scale of turbulence
33 $Le$	Lewis number
34 $P$	probability density function (PDF)
35 $P_\beta$	beta function PDF

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36	$Re_t$	turbulent Reynolds number
37	$S_{ij}$	components of the rate-of-strain tensor
38	$S_L$	laminar flame speed
39	$T$	temperature
40	$t$	time
41	$U_t$	turbulent burning velocity
42	$\mathbf{u} = \{u_1, u_2, u_3\}$	velocity vector
43	$u'$	rms turbulent velocity
44	$\mathbf{W} = \{W_1, \dots, W_N\}$	rates of production/consumption of species $n = 1, \dots, N$
45	$\mathbf{x} = \{x_1, x_2, x_3\}$	spatial coordinates
46	$x$	coordinate axis normal to the mean flame brush
47	$\mathbf{Y} = \{Y_1, \dots, Y_N\}$	mass fractions of species $n = 1, \dots, N$
48	$\delta_L = (T_b - T_u)/\max \nabla T $	laminar flame thickness
49	$\varepsilon$	dissipation rate
50	$\eta$	Kolmogorov length scale
51	$\Lambda$	width of computational domain
52	$\nu$	kinematic viscosity
53	$\xi$	sample variable
54	$\rho$	density
55	$\tau_t = L_{xx}/u'$	eddy turn over time
56	$\Phi$	equivalence ratio
57	<i>Subscripts</i>	
58	$b$	burned
59	$c$	combustion progress variable
60	$F$	fuel
61	$L$	laminar
62	$T$	temperature
63	$t$	turbulent
64	$u$	unburned
65	<i>Operators</i>	
66	$\bar{\cdot}$	Reynolds-averaged quantity
67	$\tilde{\cdot}$	Favre-averaged quantity
68	$\langle \cdot \rangle$	quantity averaged over a transverse plane

## 69 I. INTRODUCTION

70 Turbulent burning is a highly non-linear multiscale phenomenon, which involves a number of bulk and local effects to be  
71 explored. Accordingly, several alternative methods are developed and adopted to model the influence of turbulence on  
72 combustion today. One of the most promising approaches, whose development Prof. E.E. O'Brien contributed<sup>1-10</sup> significantly  
73 to, deals with a transport equation for a Probability Density Function (PDF) of a single scalar characteristic of the mixture state  
74 in a flame. Significant progress made in research into the PDF transport equation is reviewed elsewhere.<sup>11-14</sup> In particular, this  
75 approach (i) allows researchers to easily solve the problem of averaging the rate of product creation, while this problem is the  
76 major challenge to alternative models of turbulent burning, and (ii) can directly be applied to various types of flames (premixed,  
77 non-premixed, or partially premixed). In the following, solely premixed burning is addressed and the so-called combustion  
78 progress variable  $c$ , which varies from zero in fresh reactants to unity in combustion products, is considered to be a single scalar  
79 characteristic of the mixture state in an adiabatic, iso-baric, equidiffusive, and single-step chemistry flame.

80 In addition to the classical problem of predicting the PDF  $P(c)$ , recent trends in R&D of ultra-clean and highly efficient  
81 combustion technologies pose new challenges for modeling. In particular, due to strict legislation on emissions from engines,  
82 the problem of predicting concentrations of various species (not only reactants and major products, but also intermediate species  
83 such as CO, CH<sub>2</sub>O, O, H, OH, etc.) in turbulent flames has been attracting a growing attention. To average concentrations of  
84 various species using a PDF  $P(c)$ , which is either obtained by solving an appropriately closed transport equation or is modeled  
85 in another way, dependencies of the local species concentrations on  $c$  should also be invoked. For this purpose, the so-called  
86 flamelet concept<sup>15</sup> is widely used, e.g. see Table 4 in a review paper by Gicquel et al.<sup>16</sup> or Tables 5 and 6 in on a review paper  
87 by Lipatnikov.<sup>17</sup> The concept assumes adopting results (the so-called flamelet library) of numerical simulations of a set of  
88 laminar premixed flames (representative of local inherently laminar flamelets in a turbulent flow), performed by invoking an  
89 appropriately detailed model of molecular transport and chemical mechanism. Using an available technique such as Flamelet  
90 Prolongation of Intrinsic Low-Dimensional Manifolds<sup>18</sup> (FPI) or Flamelet Generation Manifold<sup>19</sup> (FGM), these results can be  
91 stored in a form of dependencies of temperature  $T_L(c)$ , density  $\rho_L(c)$ , mass fractions  $Y_{n,L}(c)$  and mass rates  $W_{n,L}(c)$  of  
92 consumption/production of  $n = 1, \dots, N$  species on the combustion progress variable  $c$ .<sup>20</sup> Finally, the following Reynolds-  
93 averaged equations

$$\bar{\mathbf{W}}(\mathbf{x}, t) = \int_0^1 \mathbf{W}_L(c) P(c, \mathbf{x}, t) dc, \quad (1)$$

$$\bar{\mathbf{Y}}(\mathbf{x}, t) = \int_0^1 \mathbf{Y}_L(c) P(c, \mathbf{x}, t) dc, \quad (2)$$

$$\bar{T}(\mathbf{x}, t) = \int_0^1 T_L(c) P(c, \mathbf{x}, t) dc, \quad (3)$$

$$\bar{\rho}(\mathbf{x}, t) = \int_0^1 \rho_L(c) P(c, \mathbf{x}, t) dc \quad (4)$$

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(or counterpart filtered equations for Large Eddy Simulation, LES) are applied to evaluate the mean (or filtered, respectively) production/consumption rates  $\bar{\mathbf{W}}$ , mass fractions  $\bar{\mathbf{Y}}$ , temperature  $\bar{T}$ , and density  $\bar{\rho}$ , respectively. Here,  $\mathbf{W}$  and  $\mathbf{Y}$  are  $N$ -dimensional vector-functions that encompass reaction rates  $W_n$  and mass fractions  $Y_n$ , respectively, for  $1 \leq n \leq N$  species.

In spite of the wide use of the flamelet concept coupled with a PDF  $P(c)$  in numerical research into premixed or stratified turbulent combustion,<sup>21-45</sup> such an approach definitely requires further study. In particular, its validation has yet been mainly performed in *a posteriori* RANS<sup>21,26,27,30,32,40</sup> or LES<sup>22,24,25,28,29,31,33-37,39,41-45</sup> studies, with the reported results showing limited capabilities of the approach for predicting mean concentrations of intermediate species such as (i) CO, e.g., see Fig. 24 in a paper by Galpin et al.,<sup>24</sup> Fig. 18 in a paper by Kolla and Swaminathan,<sup>27</sup> Figs. 9 and 13 in a paper by Lecocq et al.,<sup>29</sup> Fig. 10 in a paper by Darbyshire and Swaminathan,<sup>30</sup> Fig. 25 in a paper by Nambully et al.,<sup>35</sup> Fig. 9 in a paper by Nambully et al.,<sup>36</sup> Fig. 20 in a paper by Langella et al.,<sup>41</sup> or Fig. 18 in a paper by Donini et al.,<sup>43</sup> (ii) OH, e.g., see Fig. 11 in a paper by Langella and Swaminathan<sup>39</sup> or Fig. 21 in a paper by Langella et al.,<sup>41</sup> (iii) H<sub>2</sub> in hydrocarbon-air flames, e.g., see Fig. 10 in a paper by Darbyshire and Swaminathan,<sup>30</sup> Fig. 12 in a paper by Langella and Swaminathan,<sup>39</sup> or Fig. 20 in a paper by Langella et al.,<sup>41</sup> and (iv) CH<sub>2</sub>O, e.g., see Fig. 5 in a paper by Galeazzo et al.<sup>44</sup> However, these results are not sufficient to draw the negative conclusion regarding the flamelet concept. Indeed, first, predictive capabilities of Eq. (1) and Eqs. (2)-(4) can be significantly different, as will be discussed later. Second, substantial disagreement between computed (RANS or LES) and measured or Direct Numerical Simulation (DNS) data, observed in the aforementioned figures, could stem not only from eventual limitations of the flamelet concept, but also from limitations of the invoked PDFs, as well as other models adopted in a *a posteriori* study. For instance, as reviewed earlier,<sup>46,47</sup> capabilities of available models for predicting thermal expansion effects in premixed turbulent flames are limited and such limitations could account for the disagreement discussed here.

Therefore, there is need for *a priori* study that allows us to assess predictive capabilities of Eqs. (1)-(4) under various conditions. Such an assessment appears to be of interest, because recent experimental and DNS data reviewed by Driscoll et al.<sup>48</sup> indicate that the domain of the flamelet concept validity is substantially wider than it was earlier assumed. This hypothesis results from comparison of profiles of conditioned quantities extracted from highly turbulent flames with the counterpart profiles obtained from laminar flames,<sup>48</sup> see also recent experimental data by Skiba et al.<sup>49</sup> The hypothesis implies that Eqs. (1)-(4) could perform well even in sufficiently intense turbulence. However, *a priori* quantitative assessment of Eqs. (1)-(4) has so far been very limited. In particular, Domingo et al.<sup>22</sup> demonstrated that Eq. (2) could predict filtered mass fraction of OH, extracted from their two-dimensional DNS data obtained from a weakly turbulent (turbulent Reynolds number was as low as 55) flame at a single distance from flame-holder. Moreover, Lapointe and Blanquart<sup>42</sup> analyzed their DNS data to *a priori* explore Eq. (1) applied to a single rate  $\bar{W}_c$  of product creation (i.e., the source term in the transport equation for  $\bar{c}$ ).

Recently, two of the present authors<sup>50-53</sup> (i) analysed DNS data obtained by Dave and Chaudhuri<sup>54</sup> and by Im et al.<sup>55-59</sup> from lean complex-chemistry hydrogen-air turbulent flames characterized by different Karlovitz numbers and (ii) quantitatively validated Eqs. (2)-(4) not only for major reactants H<sub>2</sub> and O<sub>2</sub>, product H<sub>2</sub>O, temperature, and density, but also for the radicals H, O, and OH by adopting actual PDFs  $P(c)$  extracted from the same DNS data. In line with other recent data reviewed by Driscoll et al.,<sup>48</sup> these numerical findings indicate that the flamelet approach could be useful even under highly turbulent

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conditions, and, therefore, call for further assessment of Eqs. (1)-(4) for other fuels and in more intense turbulence. The present work responds to this request by performing *a priori* quantitative assessment of Eqs. (1)-(4) for various species using recent DNS data<sup>45,60,61</sup> obtained from lean methane-air flames under conditions of moderate, intense, and very intense turbulence. This is the major goal of the present study. It is worth stressing again that, with the exception of the aforementioned papers by Domingo et al.<sup>22</sup> and Lipatnikov et al.,<sup>50-53</sup> the present authors are not aware of another investigation aimed at *a priori* quantitative assessment of Eq. (2) for intermediate species such as CO or the radicals H, O, OH, etc. in premixed or stratified turbulent flames. In particular, the present authors are not aware of *a priori* quantitative assessment of Eq. (2) for intermediate species against results of a 3D complex-chemistry DNS of a  $C_8H_8$ -air flame or a flame characterized by  $Ka$  significantly larger than 100.

The present work is not limited to exploring Eqs. (1)-(4) all together but aims also at testing each equation separately. Indeed, while both Eq. (1) and Eqs. (2)-(4) stem from the same flamelet concept, the latter equations could perform better in a turbulent flow, because variations in the mass fractions  $Y_n$ , temperature  $T$ , or density  $\rho$  in a flame are smoother than variations in the rates  $W_n$ . Accordingly, eventual errors associated with the flamelet concept, i.e. reduction of  $Y_n(\mathbf{x}, t)$  and  $W_n(\mathbf{x}, t)$  to  $Y_{n,L}[c(\mathbf{x}, t)]$  and  $W_{n,L}[c(\mathbf{x}, t)]$ , respectively, and eventual errors in modeling  $P(c)$  could result in significantly larger errors in averaging the rates  $W_n$  when compared to averaging the mass fractions  $Y_n$ . This was indeed shown recently.<sup>50-53</sup>

Note that, in spite of their apparent similarity, Eqs. (1) and (2) aim at solving basically different problems, i.e. prediction of the mean rate  $\bar{W}_c$  of product creation and evaluation of mean mass fractions of various species. Accordingly, hypotheses and models developed to solve the former problem, which was also attacked in many studies that did not invoke Eq. (1), may differ significantly from hypotheses and models developed to solve the latter problem. The present focus is mainly placed on the latter problem, i.e. evaluation of mean mass fractions of various species adopting Eq. (2).

In addition to the major goal stated above, i.e. separately testing the flamelet Eq. (1) and Eqs. (2)-(4), the present work aims also at assessing the so-called presumed PDF approach. While a PDF for the combustion progress variable can be found by solving an appropriately closed transport equation<sup>4,11-14</sup> for  $P(c, \mathbf{x}, t)$ , another option known as a presumed PDF approach is commonly taken in applied CFD research into turbulent flames due to its computational efficiency. That approach consists in<sup>62-64</sup> (i) assuming a general shape  $P(c)$  of the PDF, which still involves a few unknown parameters, and (ii) evaluating these parameters by comparing values of the first moments of the  $c(\mathbf{x}, t)$ -field, calculated using the PDF, with the values of these moments, obtained by solving appropriately closed transport equations, e.g. for the Reynolds-averaged  $\bar{c}(\mathbf{x}, t)$  or the Favre-averaged  $\bar{c}(\mathbf{x}, t) \equiv \bar{\rho} \bar{c}(\mathbf{x}, t) / \bar{\rho}(\mathbf{x}, t)$  and  $\overline{c^2}(\mathbf{x}, t)$  or  $\overline{c^2}(\mathbf{x}, t) \equiv \overline{\rho c^2}(\mathbf{x}, t) / \bar{\rho}(\mathbf{x}, t)$ , respectively. More specifically, (i) the mean source terms  $\bar{W}_c(\mathbf{x}, t)$  and  $\overline{cW}_c(\mathbf{x}, t)$  in the transport equations for  $\bar{c}(\mathbf{x}, t)$  and  $\overline{c^2}(\mathbf{x}, t)$ , respectively, are closed invoking the presumed PDF, (ii) the transport equations are numerically integrated, (iii) the PDF parameters are recalculated using the obtained fields of  $\bar{c}(\mathbf{x}, t)$  and  $\overline{c^2}(\mathbf{x}, t)$ , and, finally, (iv) Eq. (2) is applied to evaluate mean concentrations of various species.

The PDF shape can be presumed adopting a sum of Dirac delta functions,<sup>65</sup> various combinations of Dirac delta functions and a flamelet PDF,<sup>22,23,26,66-68</sup> or the following beta function<sup>62-64</sup>

$$P_{\beta}(c, \bar{c}, \bar{c}^2) = \frac{\Gamma(a+b)}{\Gamma(a)\Gamma(b)} c^{a-1} (1-c)^{b-1}, \quad (5)$$

$$a = \bar{c} \left( \frac{1}{g} - 1 \right), \quad b = (1 - \bar{c}) \left( \frac{1}{g} - 1 \right). \quad (6)$$

Here,  $g = \bar{c}^2 / [\bar{c}(1 - \bar{c})]$  is the segregation factor,  $\bar{c}^2 = \overline{c^2} - \bar{c}^2$  is the variance of  $c$ , and the gamma function  $\Gamma(a) = \int_0^\infty \zeta^{a-1} e^{-\zeta} d\zeta$  is required to satisfy the normalization constraint of  $\int_0^1 P(c) dc = 1$ . Henceforth, dependencies of  $\bar{c}$ ,  $\bar{c}^2$ ,  $g$ ,  $a$ ,  $b$ , etc. on  $\mathbf{x}$  and  $t$  are not specified for brevity. Equations similar to Eqs. (5) and (6) can also be written using mass-weighted PDF  $\tilde{P}_{\beta}(c, \bar{c}, \bar{c}^2) = \rho(c) P_{\beta}(c, \bar{c}, \bar{c}^2) / \bar{\rho}$  and the Favre-averaged first,  $\bar{c}$ , and second,  $\bar{c}^2$ , moments. The latter option is often preferred, because, formally, transport equations for  $\bar{c}$  and  $\bar{c}^2$  involve a smaller number of unclosed terms than those for  $\bar{c}$  and  $\bar{c}^2$ . In the present paper, the former option is taken, because the use of direct statistics or mass-weighted statistics in Eqs. (1)-(6) is equally justified from the fundamental perspective and results reported in the following are basically similar for both statistics. In applied CFD research, the presumed beta-function PDF is widely accepted, because its shape is very flexible and, depending on the values of  $a$  and  $b$ , the PDF  $P_{\beta}(c, \bar{c}, \bar{c}^2)$  can vary from a quasi-bi-modal PDF ( $g \rightarrow 1$ ) associated with the flamelet regime of premixed turbulent combustion<sup>69</sup> to a quasi-Gaussian PDF ( $g \ll 1$ ) associated with extreme turbulence (or with a small filter size in the case of LES). Moreover, the numerical efficiency of the approach benefits from the simple algebraic relations given by Eq. (6).

Accordingly, a secondary goal of the present work consists in assessing the presumed beta-function PDF approach against the DNS data.<sup>45,60,61</sup> In addition to the aforementioned major and secondary goals, the work aims also at exploring different choices of combustion progress variable.

In the next section, DNS data analyzed for these purposes are briefly summarized. The test results are reported in Section III, with implications of these results for modeling being discussed in Section IV. Conclusions are drawn in Section V.

## II. DIRECT NUMERICAL SIMULATION

Since the DNS are discussed in detail elsewhere,<sup>45</sup> see cases A1, A2, and A3 therein, we will restrict ourselves to a brief summary of the simulations. They dealt with statistically 1D, planar premixed flames that propagated from right to left along the  $x$ -axis in a rectangular box ( $2\Lambda \times \Lambda \times \Lambda$ ) discretized on a uniform mesh of  $2N \times N \times N$  nodes. The periodic and convective outflow boundary conditions were set on the transverse sides and the outlet, respectively. To keep a flame near the domain center, the mean inlet velocity was adjusted to match the flame speed. Homogeneous, isotropic, statistically stationary turbulence was pre-generated using forcing in a cube with the periodic boundary conditions. This pre-generated turbulence was used to set the initial conditions. The same (statistically) turbulence entered the computational domain through the left boundary during combustion simulations. Inside the domain, the turbulence was forced adapting a method discussed elsewhere.<sup>70,71</sup>

At  $t = 0$ , a planar laminar flame ( $\text{CH}_4$ -air mixture with the equivalence ratio  $\Phi = 0.6$  under the atmospheric conditions, the laminar flame speed  $S_L = 0.12$  m/s and thickness  $\delta_L = (T_b - T_u) / \max|\nabla T| = 0.92$  mm) was embedded into the computational domain at  $x = \Lambda$ . The continuity, low-Mach-number Navier-Stokes, species and energy transport equations were

numerically solved. A skeletal mechanism (16 species and 35 reactions) by Smooke and Giovangigli<sup>72</sup> was used. Differential diffusion effects and temperature-dependence of molecular transport coefficients were modeled using Fourier's and Fick's laws with mixture-averaged transport properties calculated following CHEMKIN. Soret and Dufour effects were neglected.

The DNS solver was described in detail and validated elsewhere.<sup>73</sup> A 5th order weighted essentially non-oscillatory (WENO) finite difference method was used for convective terms and a 6th order central difference scheme was used for all other terms. For unsteady terms, a second-order operator splitting scheme<sup>74</sup> was adopted by integrating chemical source terms between two half-time-step integrations of the diffusion term. The integration of the diffusion term was further divided into smaller explicit steps to ensure stability. The overall time step was set to get the CFL number smaller than 0.1. Reaction rates in species transport equations were integrated using the stiff DVODE solver.<sup>75</sup> The variable-coefficient Poisson equation for pressure differences was solved adopting a multigrid method.<sup>76</sup>

**Table I. Simulation conditions**

case	$N$	$L_{xx}/\delta_{th}$	$\eta/\delta_{th}$	$\delta_{th}/\Delta x$	$u'/S_L$	$Ka$	$Da$	$Re_t$
A1	256	1.3	0.105	23.5	3.7	6.0	0.38	32
A2	256	1.0	0.036	23.5	18.	74.	0.06	120
A3	512	1.0	0.021	47.0	66.	540.	0.015	390

Three cases characterized by different rms velocities  $u'$  and, hence, different Karlovitz numbers  $Ka = (u'/S_L)^{3/2}(\delta_{th}/L_{xx})^{1/2}$ , Damköhler numbers  $Da = L_{xx}S_L/(u'\delta_{th})$ , and turbulent Reynolds numbers  $Re_t = u'L_{xx}/\nu_u$ , see Table I, were simulated. Here,  $L_{xx}$  is the axial longitudinal integral length scale evaluated by integrating the correlation function for the axial velocity;  $\eta = (\nu^3/\epsilon)^{1/4}$  is the Kolmogorov length scale;  $\nu_u$  is the kinematic viscosity of unburned mixture;  $\epsilon = 2\nu S_{ij}S_{ij}$  is the rate of dissipation of turbulent kinetic energy;  $S_{ij} = (\partial u_i/\partial x_j + \partial u_j/\partial x_i)/2$  is the rate-of-strain tensor;  $\Delta x = \Delta y = \Delta z$  is the grid spacing; the summation convention applies to repeated indexes; and all turbulence characteristics are averaged over the volume of a cube where the turbulence is pre-generated. The computational domain width is  $\Lambda = 5$  mm.

Six different combustion progress variables are defined as follows  $c_k = (\phi_k - \phi_{k,u})/(\phi_{k,b} - \phi_{k,u})$ , where  $\phi_1 = Y_{CH_4}$ ,  $\phi_2 = Y_{O_2}$ ,  $\phi_3 = Y_{H_2O}$ , and  $\phi_4 = Y_{CO_2}$ , are the mass fractions of  $CH_4$ ,  $O_2$ ,  $H_2O$ , and  $CO_2$ , respectively,  $\phi_5 = Y_{CO_2} + Y_{CO}$ , and  $\phi_6 = T$ . Note that the dependencies of combustion progress variables defined using sums of  $Y_{H_2O} + Y_{CO_2} + Y_{CO}$  or  $Y_{H_2O} + Y_{CO_2} + Y_{CO} + Y_{H_2}$  on the temperature-based  $c_T \equiv c_6$  are almost identical to  $c_2(c_T)$  in the considered unperturbed laminar premixed flame. Accordingly, these sums were not addressed in the present study.

Mean profiles  $\bar{q}(\bar{c}_k)$  of various quantities  $q$  were evaluated as follows. First,  $q(\mathbf{x}, t)$  and  $c_k(\mathbf{x}, t)$ -fields were averaged over each transverse plane  $x = \text{const}$  at each instant  $t$  (25, 21 and 30 snapshots separated by  $\Delta t = 0.5\tau_t = 0.5 L_{xx}/u'$  in cases A1, A2 and A3, respectively). Second, the obtained profiles of  $\langle q \rangle(x, t)$  were transformed to  $\langle q \rangle(\bar{\xi})$  using the profiles of  $\langle c_k \rangle(x, t)$  divided into 51 intervals. Here,  $\bar{\xi}$  is a sample variable for  $\langle c_k \rangle(x, t)$  and a transverse plane  $x = \text{const}$  contributes to the value of  $\langle q \rangle(\bar{\xi}_j)$  if  $|\langle c_k \rangle(x, t) - \bar{\xi}_j| < 0.01$  ( $\bar{\xi}_j = 0.02j$ ;  $j = 0, \dots, 50$ ). The analyzed snapshots were stored at  $t > 10\tau_t$ .

To examine Eqs. (1)-(4), the DNS PDFs  $P_k(\xi, x, t)$  were sampled from grid points characterized by  $|c_k(\mathbf{x}, t) - \xi_j| < 0.0025$  ( $\xi_j = 0.005j$ ;  $j = 0, \dots, 200$ ) in each transverse plane  $x = \text{const}$  at each instant  $t$ . Here,  $\xi$  is a sample variable for the



instantaneous  $c_k(x, t)$ -fields. Subsequently, the PDFs  $P_k(\xi, x, t)$  were transformed to  $P_k(\xi, \bar{\xi})$  using the profiles of  $\langle c_k \rangle(x, t)$ , as discussed above. To assess the presumed  $\beta$ -function PDF approach, the first,  $\langle c_k \rangle(x, t)$  or  $\bar{c}_k(x)$ , and second,  $\langle c_k^2 \rangle(x, t)$  or  $\bar{c}_k^2(x)$ , respectively, moments extracted from the DNS data were substituted into Eq. (6), followed by substitution of the obtained values of  $a$  and  $b$  into Eq. (5). Finally, six sets of dependencies of  $\bar{W}(\bar{c}_k)$ ,  $\bar{Y}(\bar{c}_k)$ ,  $\bar{T}(\bar{c}_k)$ , or  $\bar{\rho}(\bar{c}_k)$  were computed for the six  $c_k$  using the two types of PDFs and Eq. (1), (2), (3), or (4), respectively.

### III. RESULTS AND DISCUSSION

Figure 1 shows that, in all three cases, Eq. (3) very well predicts the mean temperature using both the actual and presumed  $\beta$ -function PDFs and adopting the oxygen-based combustion progress variable  $c_2$  or, to a lesser extent, the water-based  $c_3$  (dependencies of  $\bar{T}$  on the temperature-based  $\bar{c}_6$  reduce to a straight line and, therefore, are not shown). The use of the fuel-based  $c_1$  results in underestimating the mean temperature at  $\bar{c}_1 > 0.8$ . Worst predictions are obtained adopting  $c_4$  and  $c_5$ , which both are based on the mass fraction of  $\text{CO}_2$ . These differences between mean temperatures extracted from the DNS data and yielded by Eq. (3) will be discussed later. The dependencies  $\bar{T}(\bar{c}_k)$  calculated using the actual and presumed  $\beta$ -function PDFs are hardly distinguishable in all cases.

Figure 2 also supports the flamelet concept by quantitatively validating Eq. (4) with either the actual or the presumed  $\beta$ -function PDF for the temperature-based  $c_6$  or the fuel-based  $c_1$ . The use of  $c_2$  ( $c_3$ ) based on the mass fraction of  $\text{O}_2$  ( $\text{H}_2\text{O}$ , respectively) yields slightly underestimated (overestimated, respectively)  $\bar{\rho}$  in cases A1 and A2 (A2 and A3, respectively), but the differences are rather small at least for  $c_2$ . Similar to Fig. 1, worst predictions are obtained adopting  $c_4$  and  $c_5$ . It is of interest to note that while the flamelet Eq. (4) performs well under conditions of the present study, the computed dependencies  $\bar{\rho}(\bar{c}_k)$  are non-linear contrary to the well-known Bray-Moss-Libby<sup>69</sup> (BML) linear relation of  $\bar{\rho} = \rho_u(1 - \bar{c}) + \rho_b\bar{c}$ . Since the BML theory relies not only on the flamelet concept, but also (and mainly) on a hypothesis that the probability of finding intermediate states of the mixture is much less than unity, the discussed observation implies that this hypothesis does not hold under conditions of the present study. This could be expected, because  $Ka > 1$  in all three cases.

Figures 3 and 4 quantitatively validate the flamelet Eq. (2) for major reactants and products, including  $\text{CO}$ , provided that the combustion progress variable is defined using the temperature, see the bottom row, with both the actual and  $\beta$ -function PDFs yielding very good results (note that  $\bar{Y}_{\text{CO}_2}$  is slightly underestimated in cases A2 and A3). For four other  $\bar{c}_k$ , the computed results are generally good, but Eq. (2) performs worse for some species in some cases, e.g. (i) for  $\bar{Y}_{\text{CO}_2}(\bar{c}_1)$  and  $\bar{Y}_{\text{CO}}(\bar{c}_1)$  in all three cases, see red lines in the first rows in Figs. 3 and 4, respectively, (ii) for  $\bar{Y}_{\text{CH}_4}(\bar{c}_2)$  in cases A2 and A3 or for  $\bar{Y}_{\text{CO}}(\bar{c}_2)$  in cases A1 and A2, see the second row in Fig. 4, or (iii) for  $\bar{Y}_{\text{CO}_2}(\bar{c}_3)$  and  $\bar{Y}_{\text{O}_2}(\bar{c}_3)$  in all three cases, see the third row in Fig. 3, or for  $\bar{Y}_{\text{CO}}(\bar{c}_3)$  in cases A1 and A2 and  $\bar{Y}_{\text{CH}_4}(\bar{c}_3)$  in cases A2 and A3, see the third row in Fig. 4. The use of the  $\text{CO}_2$ -based  $c_4$  or  $c_5$  yields the worst results for  $\bar{Y}_{\text{H}_2\text{O}}(\bar{c}_k)$  in cases A2 and A3, see black lines in the fourth row in Fig. 3, and for  $\bar{Y}_{\text{CH}_4}(\bar{c}_k)$  in all three cases, see black lines in the fourth row in Fig. 4 or 5, respectively. Moreover,  $\bar{Y}_{\text{CO}}(\bar{c}_4)$  or  $\bar{Y}_{\text{CO}}(\bar{c}_5)$  is substantially overestimated in case A3, see red lines in the fourth row in Fig. 4 or 5, respectively.

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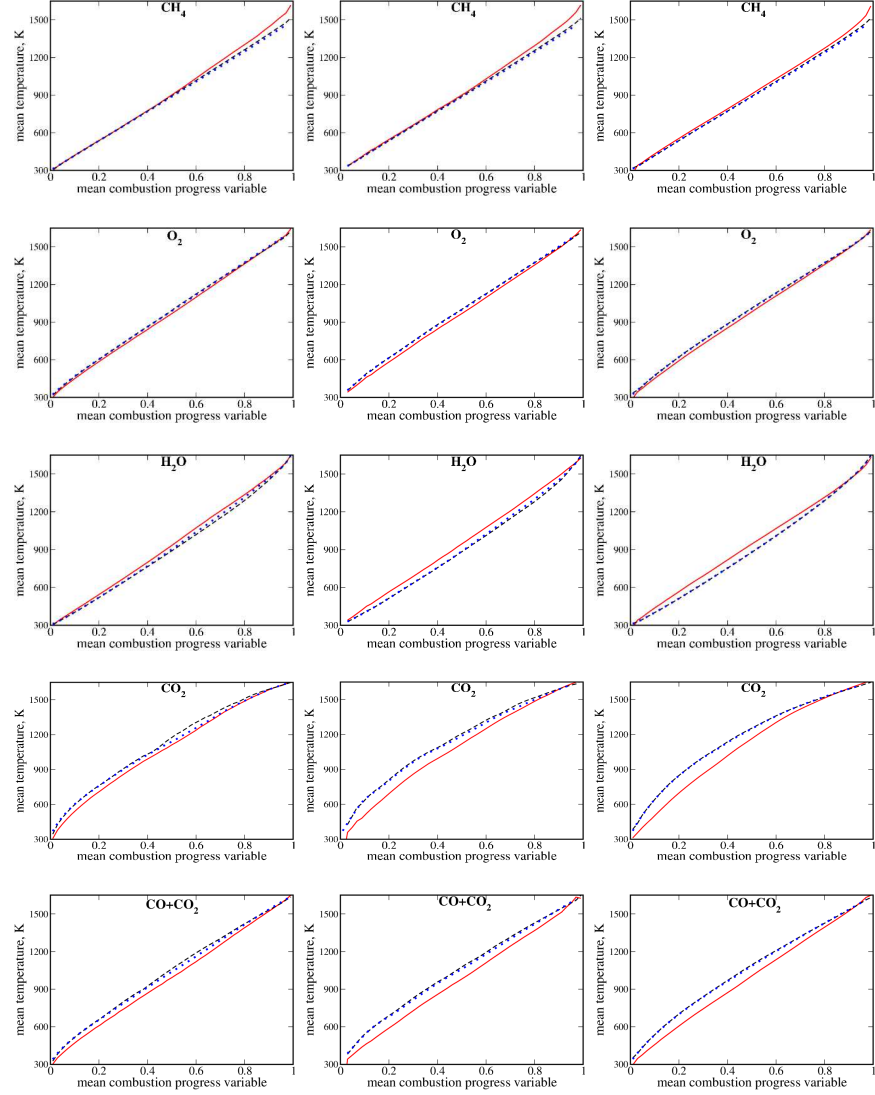
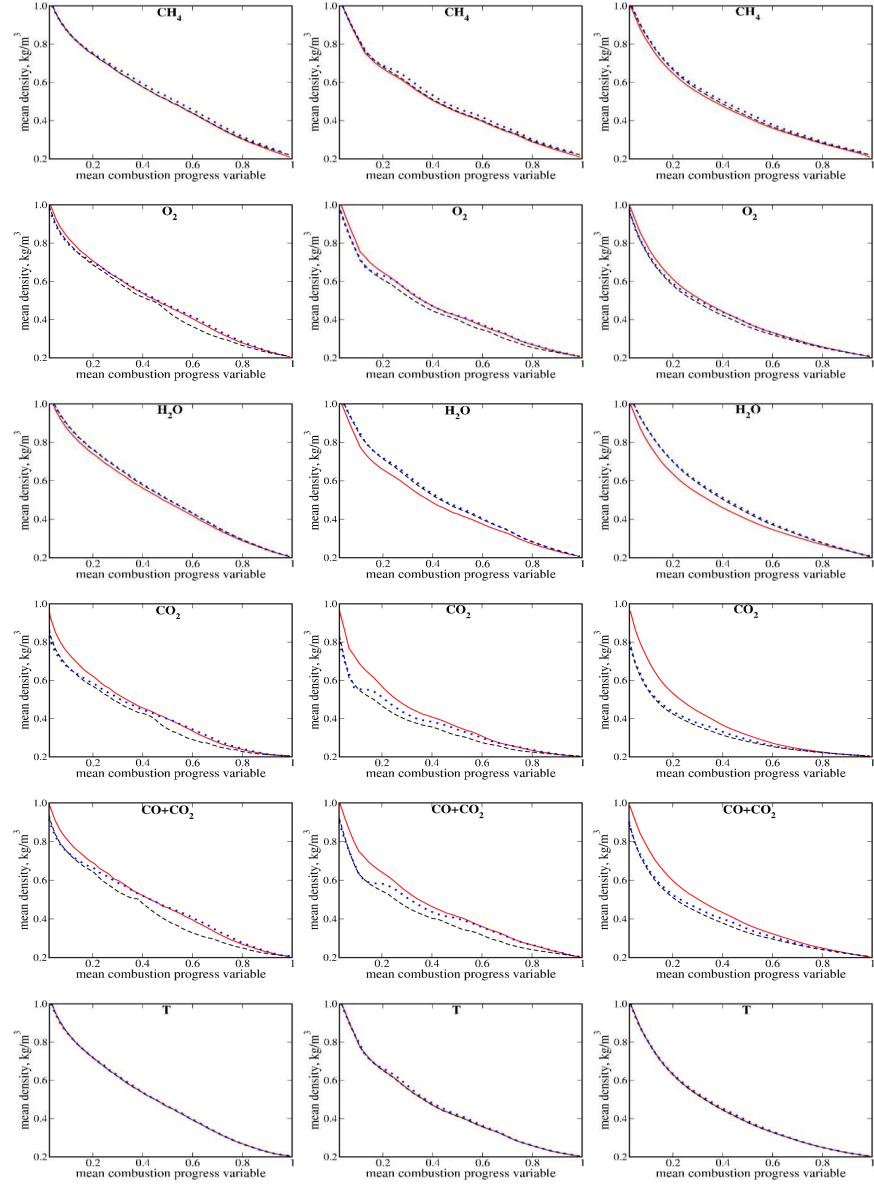


FIG. 1. Mean temperature vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Solid lines show  $\bar{T}$  extracted from the DNS data. Dashed lines show  $\bar{T}$  evaluated using the flamelet library and the PDF extracted from the DNS data. Dotted lines show  $\bar{T}$  calculated invoking the  $\beta$ -distribution PDF. Results computed in cases A1, A2, and A3 are plotted in the left, middle, and right columns, respectively.

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256 **FIG. 2.** Mean density vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Legends are  
257 explained in caption to Fig. 1.

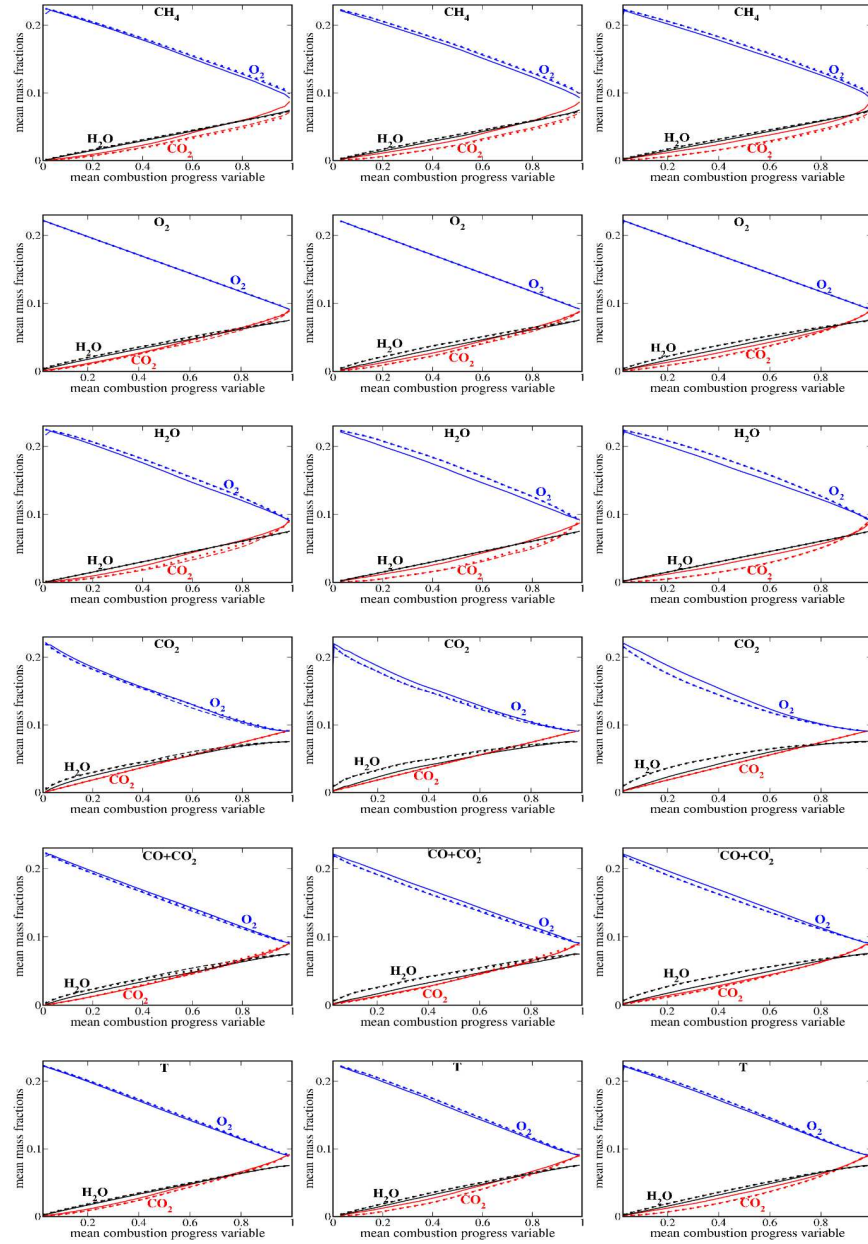


FIG. 3. Mean mass fractions of  $O_2$ ,  $H_2O$ , and  $CO_2$  vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Legends are explained in caption to Fig. 1.

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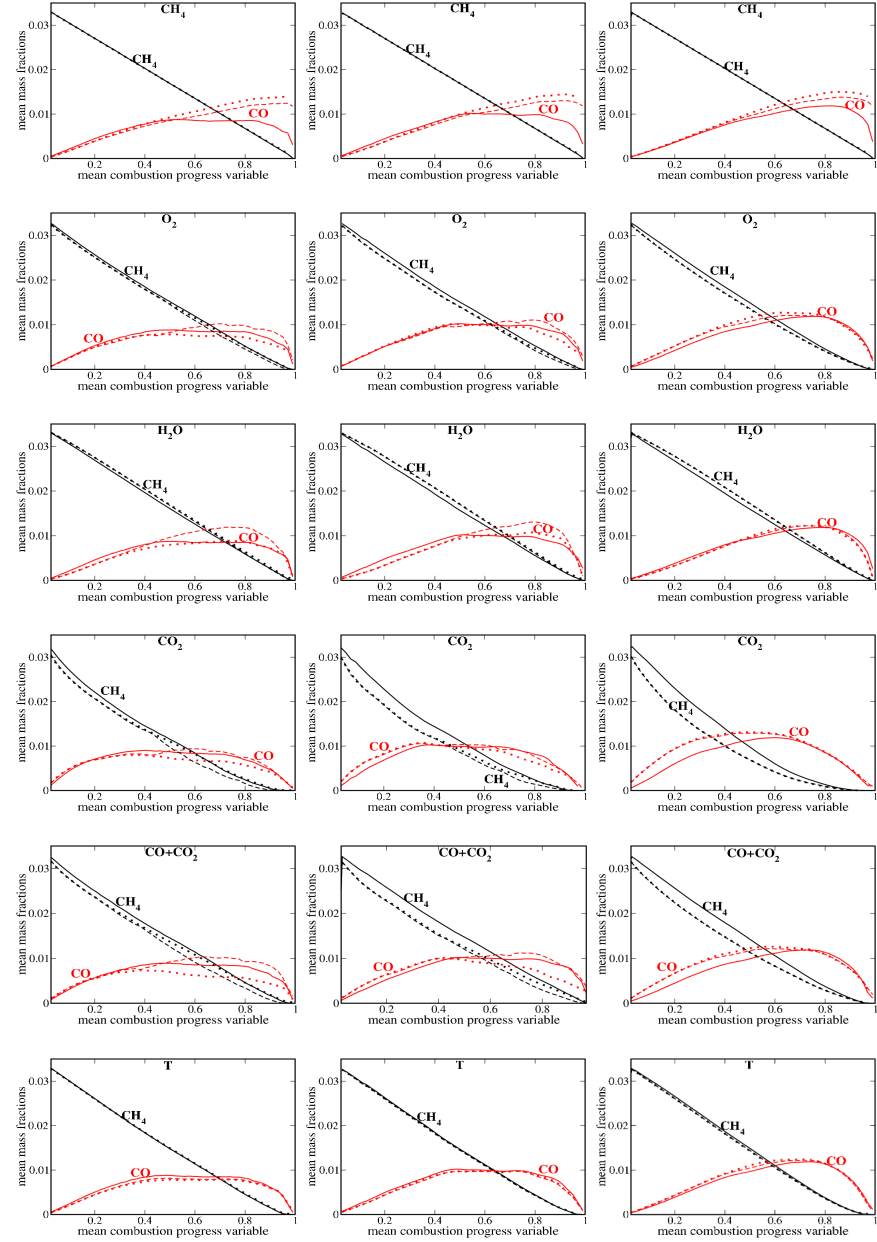


FIG. 4. Mean mass fractions of  $\text{CH}_4$  and  $\text{CO}$  vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Legends are explained in caption to Fig. 1.

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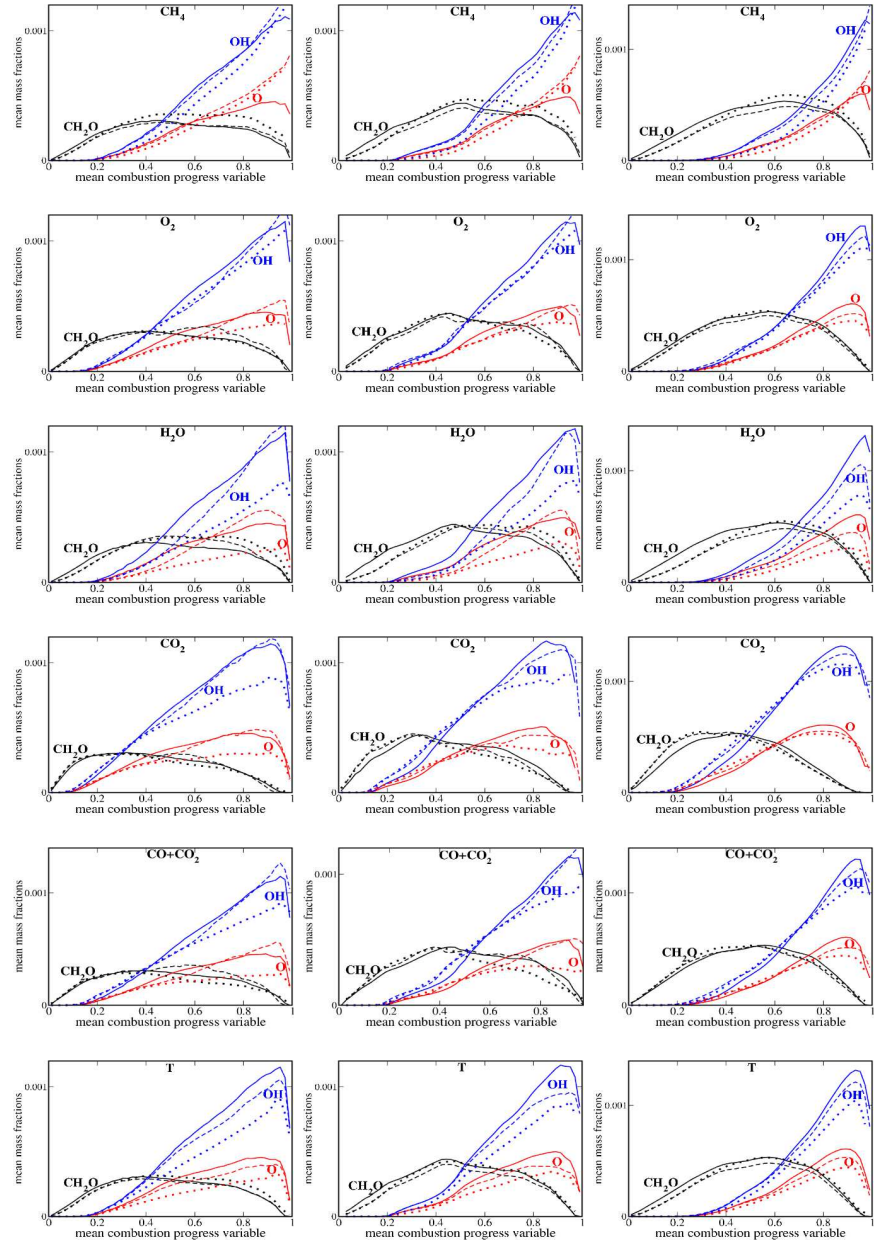


FIG. 5. Mean mass fractions of  $\text{CH}_2\text{O}$ ,  $\text{OH}$ , and  $\text{O}$  vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Legends are explained in caption to Fig. 1.

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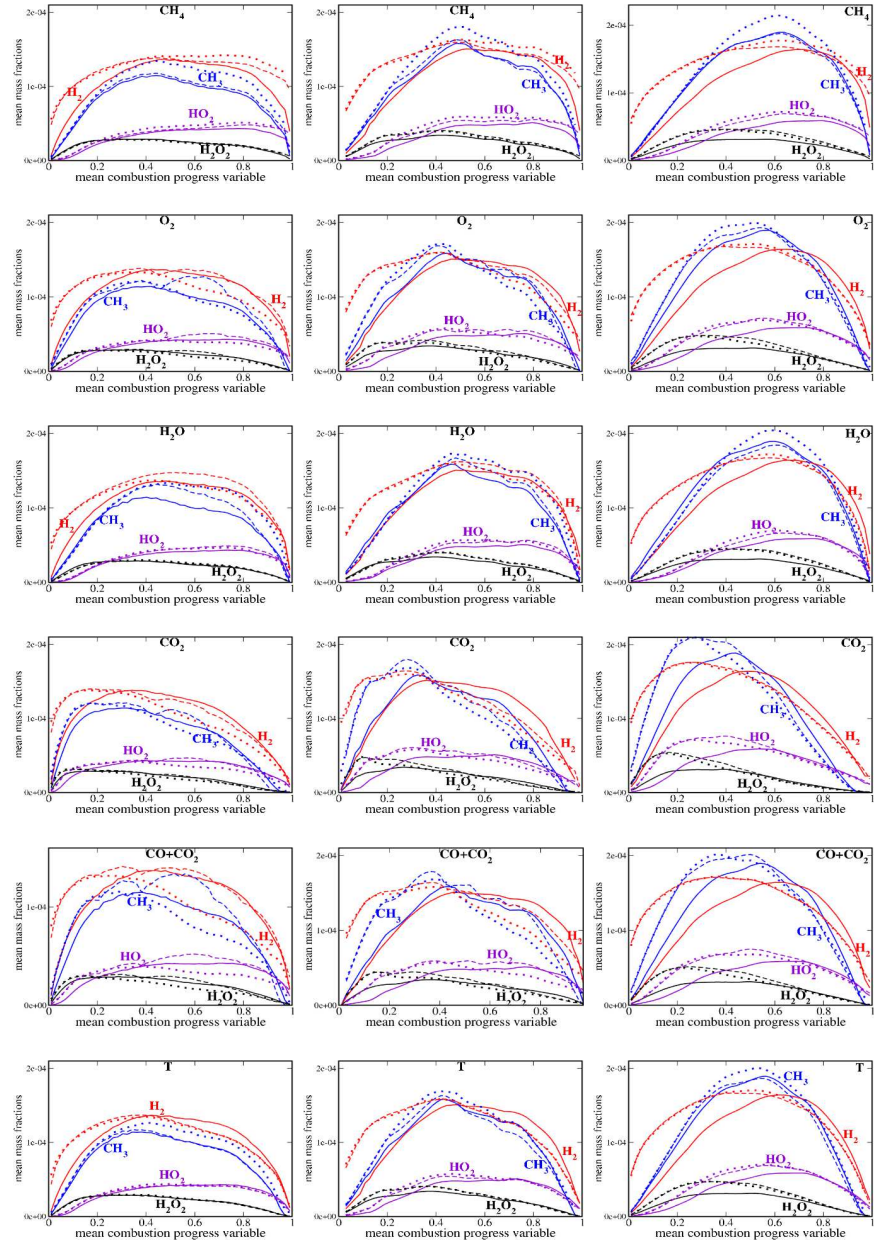
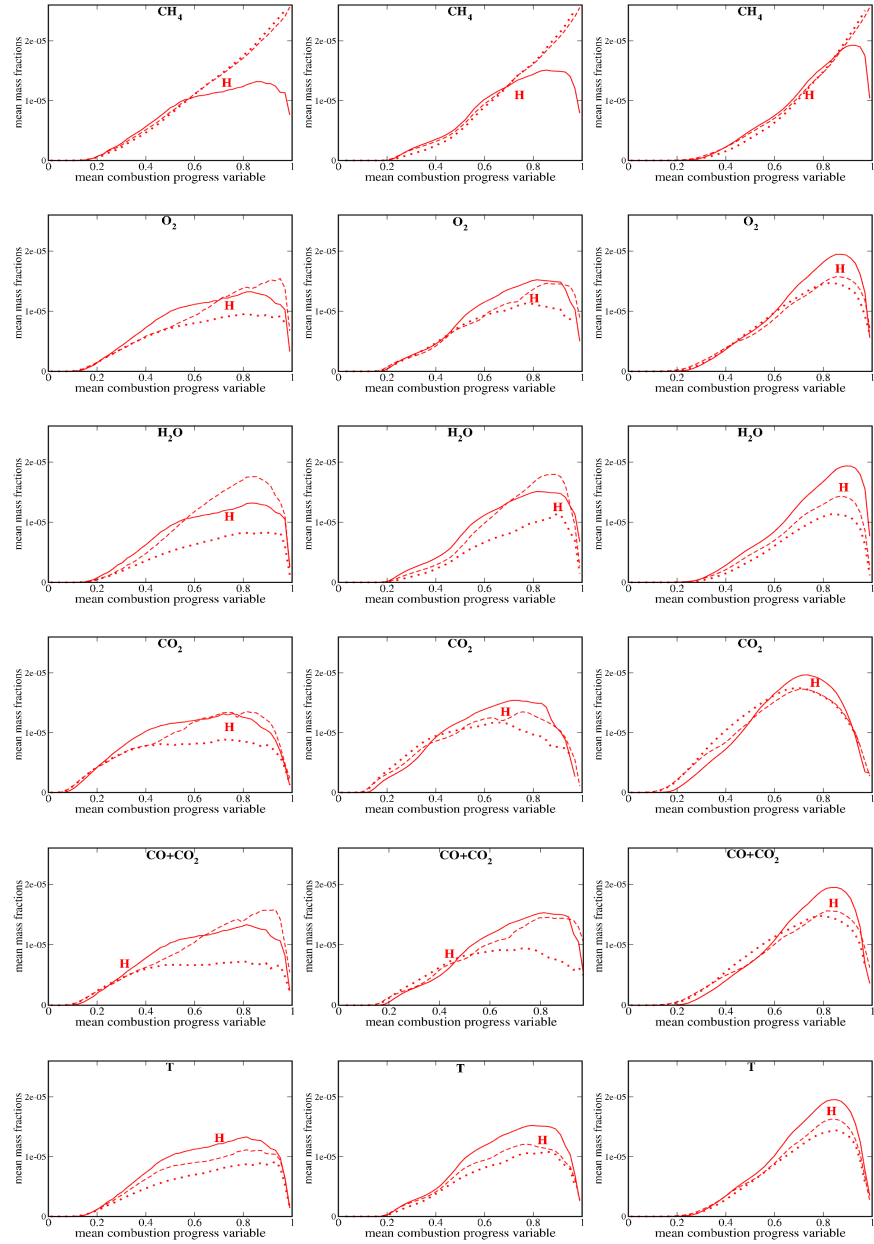


FIG. 6. Mean mass fractions of  $\text{CH}_3$ ,  $\text{H}_2$ ,  $\text{HO}_2$ , and  $\text{H}_2\text{O}_2$  vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure. Legends are explained in caption to Fig. 1.

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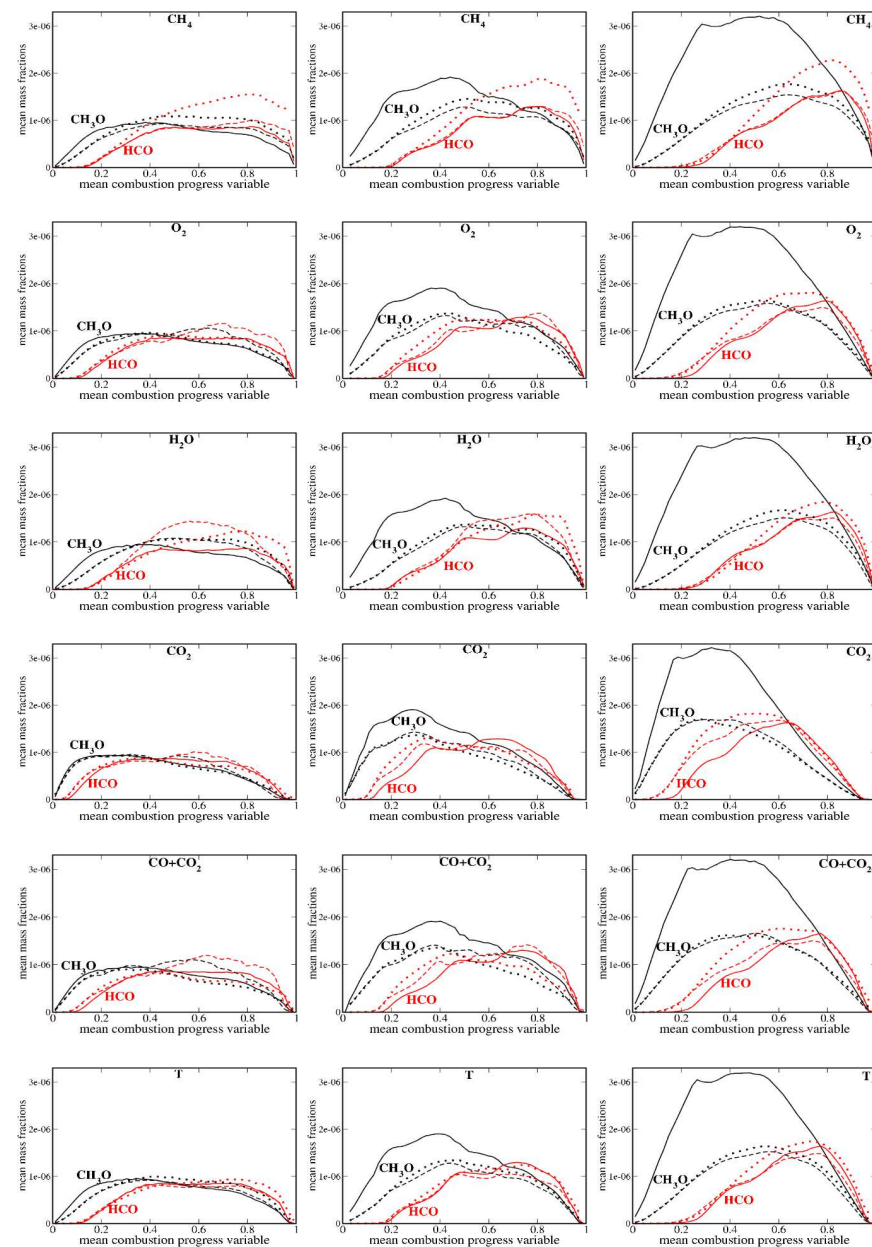


267  
268 **FIG. 7.** Mean mass fraction of H vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each subfigure.  
269 Legends are explained in caption to Fig. 1.

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270 **FIG. 8.** Mean mass fraction of  $\text{CH}_3\text{O}$  and  $\text{HCO}$  vs. various mean combustion progress variables  $\bar{c}_k$  specified in the top of each  
 271 subfigure. Legends are explained in caption to Fig. 1.

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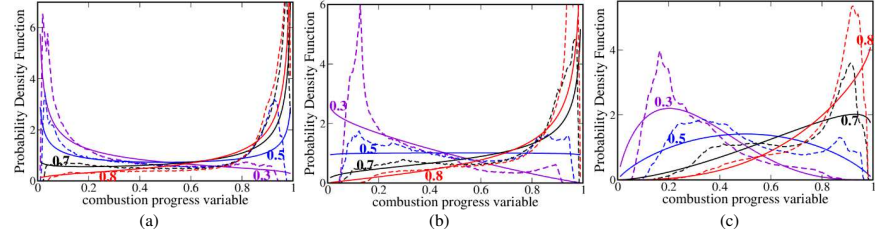
272 All in all, Figs. 1-4 quantitatively validate the flamelet Eqs. (2)-(4) at various  $6 \leq Ka \leq 540$ , at least if the combustion  
273 progress variable is defined using the temperature, with good results being obtained adopting not only the actual PDFs, but  
274 even the presumed  $\beta$ -function PDFs. However, these findings are expected, because both PDFs are built using the correct  
275 values of the first two moments of the  $c(\mathbf{x}, t)$ -field and spatial variations of the density, temperature, or mass fractions of major  
276 reactants and products are relatively smooth (weakly non-linear) in a flame. Prediction of mean mass fractions of intermediate  
277 species, whose spatial variations are substantially non-linear and are characterized by significantly smaller length scales,  
278 appears to be a much more difficult task, which is addressed for all 10 such species, considered within the framework of the  
279 skeletal mechanism by Smooke and Giovangigli,<sup>72</sup> in Figs. 5-8. The following trends are worth noting.

280 First, if (i) combustion progress variable is defined based on the temperature, as recommended above, and (ii) the PDF  
281  $P_\theta(\xi, \bar{\xi})$  is extracted from the DNS data, Eq. (2) very well predicts the mean mass fractions of  $\text{CH}_2\text{O}$ ,  $\text{CH}_3$ , and  $\text{HCO}$  in all  
282 three cases, see the bottom rows in each figure and cf. black solid and dashed lines in Fig. 5, blue solid and dashed lines in  
283 Fig. 6, and red solid and dashed lines in Fig. 8. The mean mass fractions of  $\text{OH}$  and  $\text{O}$  are slightly underestimated, cf. blue or  
284 red, respectively, solid and dashed lines in Fig. 5. The mean mass fraction of  $\text{HO}_2$  or  $\text{H}_2\text{O}_2$  is very well predicted in case A1,  
285 cf. violet or black, respectively, solid and dashed lines in Fig. 6, but Eq. (2) performs worse with increasing  $Ka$ . The mean  
286 mass fraction of  $\text{H}_2$  is overestimated at  $\bar{c}_k < c_k^*$  and the mean mass fraction of  $\text{H}$  is underestimated at  $\bar{c}_k > c_k^*$ , with  $c_k^*$  being  
287 increased with increasing  $Ka$ , cf. red solid and dashed lines in Figs. 6 and 7, respectively. At a first glance, this limitation of  
288 Eq. (2) is associated with high molecular diffusivities of  $\text{H}_2$  and  $\text{H}$ . Local phenomena caused by interaction of complex  
289 chemistry and preferential diffusion effects were already documented in DNS studies of highly turbulent lean hydrogen-air  
290 flames.<sup>77-79</sup> Finally, the mean mass fraction of  $\text{CH}_3\text{O}$  is significantly underpredicted in cases A2 and, especially, A3, cf. black  
291 solid and dashed lines in Fig. 8.

292 Second, if combustion progress variable is still defined based on the temperature, the actual and  $\beta$ -function PDFs yield  
293 almost the same results for  $\text{CH}_2\text{O}$ ,  $\text{H}_2$ ,  $\text{HO}_2$ ,  $\text{H}_2\text{O}_2$ , and  $\text{CH}_3\text{O}$ . For five other intermediate species, i.e.  $\text{OH}$ ,  $\text{O}$ ,  $\text{CH}_3$ ,  $\text{H}$ , or  $\text{HCO}$ ,  
294 the two PDFs yield substantially different results. This difference implies that Eqs. (5) and (6) do not predict the PDF extracted  
295 from the DNS data. Differences between the actual PDFs extracted from the DNS data and  $\beta$ -function PDFs built using the  
296 first two moments of the  $c_T(\mathbf{x}, t)$ -field extracted from the same DNS data are clearly seen in Fig. 9.

297 Third, in cases A2 and A3,  $\bar{Y}_n(\bar{c}_k)$  obtained by adopting the actual PDF for the oxygen-based  $c_2$  and the temperature-  
298 based  $c_5$ , are comparable for the most intermediate species, but, in case A1, the use of  $c_2$  yields substantially worse results for  
299  $\text{CO}$ , see Fig. 4,  $\text{CH}_2\text{O}$ , see Fig. 5,  $\text{CH}_3$ , see Fig. 6,  $\text{HCO}$  and  $\text{CH}_3\text{O}$ , see Fig. 8.

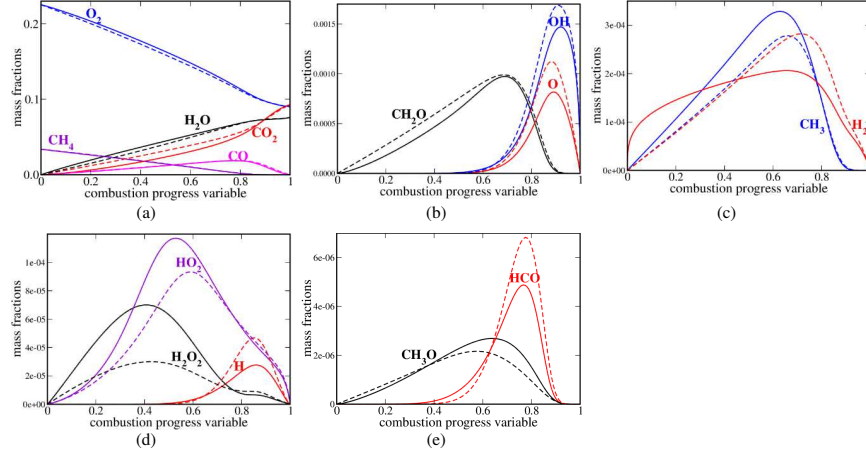
300 Fourth, even for four other combustion progress variables, results computed using Eq. (2) seem to be encouraging. For  
301 some species, the predictions are very good:  $\bar{Y}_{\text{OH}}(\bar{c}_1)$ , cf. blue solid and dashed lines in the first row in Fig. 5;  $\bar{Y}_{\text{O}}(\bar{c}_4)$ , cf. red  
302 solid and dashed lines in the fourth row in Fig. 5;  $\bar{Y}_{\text{HO}_2}(\bar{c}_3)$ , cf. violet solid and dashed lines in the third row in Fig. 6;  $\bar{Y}_{\text{CH}_3}(\bar{c}_1)$ ,  
303 cf. blue solid and dashed lines in the first row in Fig. 6;  $\bar{Y}_{\text{H}}(\bar{c}_4)$  cf. solid and dashed lines in the fourth row in Fig. 7; or  
304  $\bar{Y}_{\text{HCO}}(\bar{c}_1)$ , cf. red solid and dashed lines in the first row in Fig. 8.



**FIG. 9.** Probability density functions for the temperature-based combustion progress variable  $c_6 \equiv c_T$  obtained at  $\bar{c}_6 = 0.3$  (violet curves), 0.5 (blue curves), 0.7 (black curves), and 0.8 (red curves) from flames (a) A1 (left cell), (b) A2 (middle cell), and (c) A3 (right cell). Solid lines show  $\beta$ -function PDFs built using the first two moments of the  $c_6(\mathbf{x}, t)$ -field extracted from the DNS data. Dashed lines show actual PDFs extracted from the DNS data.

All in all, Figs. 1-8 considered all together (i) support the flamelet Eqs. (2)-(4) in a wide range of  $6 \leq Ka \leq 540$ , with certain reservations discussed above, (ii) indicate that the temperature is a better choice for defining combustion progress variable under conditions of the present study, and (iii) call for development of a better model for the combustion-progress-variable PDF. Sufficiently good quantitative agreement between the profiles of  $\bar{V}_n(\bar{c}_6)$  extracted from the DNS data and calculated using Eq. (2) with the actual PDF  $P_6(\xi, \bar{\xi})$ , obtained for almost all species at high Karlovitz numbers up to 540, is the major result of the above analysis. This result appears to be of significant importance for applied CFD research into premixed or stratified turbulent burning, because it supports the use of a simple Eq. (2) in unsteady multi-dimensional simulations. It is worth remembering, however, that such simulations invoke other models of various phenomena such as influence of turbulence on combustion, thermal expansion effects, heat losses, etc., as well as a model of the PDF  $P(c)$ . Bearing in mind good *a priori* prediction shown in Figs. 1-8, accuracy of such *a posteriori* simulation is likely to be limited by some of the aforementioned models, rather than by Eq. (2). For this reason and because the flamelet Eqs. (1)-(4) are tools for the applied CFD research, further improvement of results reported in Figs. 1-8, e.g. by invoking a flamelet library created for strained laminar premixed flames, does not seem to be of the top priority at the moment, as well as a thorough investigation of differences between the profiles of  $\bar{V}_n(\bar{c}_k)$  extracted from the DNS data and calculated using the actual PDFs  $P_k(\xi, \bar{\xi})$  extracted from the same data. Nevertheless, these differences deserve some discussion.

There are three types of such differences, which are more pronounced (i) for certain  $\bar{c}_k$  when compared to the temperature-based  $\bar{c}_6 \equiv \bar{c}_T$ , (ii) for some species even if  $\bar{c}_T$  is adopted, or (iii) in the highly turbulent flame A3. To reveal the causes of these differences, Fig. 10 shows the structure of the unperturbed laminar flame calculated using the skeletal mechanism by Smooke and Giovangigli<sup>72</sup> (solid lines), as well as the structure of the counterpart equidiffusive flame (dashed lines). In the latter case, molecular mass diffusivities of each species are equal to the molecular heat diffusivity of the mixture or, in other words,  $Le_n = 1$  for each species  $n$ . Since the use of the temperature-based combustion progress variable  $c_T$  yielded the best agreement between the DNS data and results obtained adopting Eqs. (2)-(4), Fig. 10 reports the flame structure in the  $c_T$ -space. The following trends are worth emphasizing.



**FIG. 10.** Mass fractions of (a)  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ , and  $\text{CO}$ , (b)  $\text{CH}_2\text{O}$ ,  $\text{O}$ , and  $\text{H}$ , (c)  $\text{H}_2$  and  $\text{CH}_3$ , (d)  $\text{HO}_2$ ,  $\text{H}_2\text{O}_2$ , and  $\text{H}$ , and (e)  $\text{CH}_3\text{O}$  and  $\text{HCO}$  vs. temperature-based combustion progress variable  $c_T$ . Solid and dashed lines show mass fractions calculated for unperturbed laminar flames with  $Le_n \neq 1$  and  $Le_n = 1$ , respectively.

First, Fig. 10a shows that, for  $\text{O}_2$  or  $\text{H}_2\text{O}$ , dependencies of  $Y_{n,L}(c_T)$  are almost linear at  $c_T < 0.85$ , but become weakly non-linear at larger  $c_T$ , with variations in the mass fraction of oxygen or water being less pronounced at  $c_T > 0.85$ . The same trends are also observed for the fuel, with the mass fraction of  $\text{CH}_4$  almost vanishing at  $c_T > 0.85$ . If (i) species  $k$  is selected to define a combustion progress variable  $c_k$  and (ii) the rate of change of  $Y_k$  with  $c_T$  is decreased in a certain range of  $c_T$ , i.e.  $|dY_k/dc_T|$  is small (or very small, as for  $\text{CH}_4$  at  $c_T > 0.85$ ); even small (very small for methane) variations in  $Y_k$  or  $c_k$  are accompanied with significant variations in other mass fractions  $Y_n$  in the considered range of  $c_T$ , i.e. the non-linearities of the dependencies of  $Y_{n,L}$  on  $c_k$  are more (much more for  $\text{CH}_4$ ) pronounced when compared to the non-linearity of  $Y_{n,L}(c_T)$ . For instance, the peak absolute value of the second derivative  $|d^2Y_{\text{CO}}/dc_T^2|$  reached in the unperturbed laminar flame in an interval of  $0.05 < c_T < 0.95$  is larger than the peak  $|d^2Y_{\text{CO}}/dc_T^2|$  by almost six orders of magnitude and this difference is even larger at larger  $c_T$ . Furthermore, if a dependence of  $Y_{n,L}(c_k)$  is highly non-linear, the use of  $Y_{n,L}(c_k)$  for averaging the mass fraction  $Y_n(\mathbf{x}, t)$  by adopting Eq. (2) can result in significant errors. Accordingly, differences between  $\bar{Y}_n(\bar{c}_1)$  extracted from the DNS data and calculated using Eq. (2) with the actual PDF (see the top rows in Figs. 3-8) are expected due to the highly non-linear dependencies  $Y_{n,L}(c_1)$  at large  $c_1 \equiv c_F$ , i.e. in the flame zone characterized by vanishing mass fraction of  $\text{CH}_4$  and, hence,  $c_F \approx 1$ . This effect is expected to be of the most importance at large  $\bar{c}_F$ . Such differences are well pronounced for  $\text{CO}$  (see red curves in Fig. 4),  $\text{O}$  (see red curves in Fig. 5),  $\text{H}_2$  in case A1 (see red curves in Fig. 6), and  $\text{H}$  (see Fig. 7). For  $\text{CH}_2\text{O}$ ,  $\text{CH}_3$ ,  $\text{H}_2\text{O}_2$ ,  $\text{CH}_3\text{O}$ , and  $\text{HCO}$ , such differences are weakly (if any) pronounced, because the mass fractions of these species almost vanish at large  $c_F$ , as shown in Fig. 10. The above discussion and Fig. 10 indicate that the fuel mass fraction is not the best choice for defining combustion progress variable for the studied lean methane-air flame.

Similar reasoning explains worse performance of the combustion progress variables  $c_4$  and  $c_5$ , which involve the mass fraction of  $\text{CO}_2$ . In the studied unperturbed laminar flame, the non-linearities of the dependencies  $c_4(c_T)$  and  $c_5(c_T)$  are well pronounced, with the derivatives  $dc_4/dc_T$  and  $dc_5/dc_T$  being decreased with decreasing  $c_T$ . Accordingly, at low  $c_T$ , the same variations in  $Y_n$  are accompanied with larger variations in  $c_T$  when compared to variations in  $c_4$  or  $c_5$  and the dependencies of  $Y_n(c_4)$  and  $Y_n(c_5)$  are more non-linear when compared to  $Y_n(c_T)$ . For instance, the peak absolute value of the second derivative  $|d^2\rho/dc_T^2|$  reached in the unperturbed laminar flame in an interval of  $0.05 < c_T < 0.95$  is smaller than the peak  $|d^2\rho/dc_4^2|$  or  $|d^2\rho/dc_5^2|$  by a factor of about 30 and 7.2, respectively. As a result, the use of Eq. (4) jointly with  $c_4$  or  $c_5$  yields substantially underestimated mean density, see the fourth and fifth rows in Fig. 2. An increase in the effect magnitude with  $Ka$  may be attributed to stronger fluctuations in more intense turbulence.

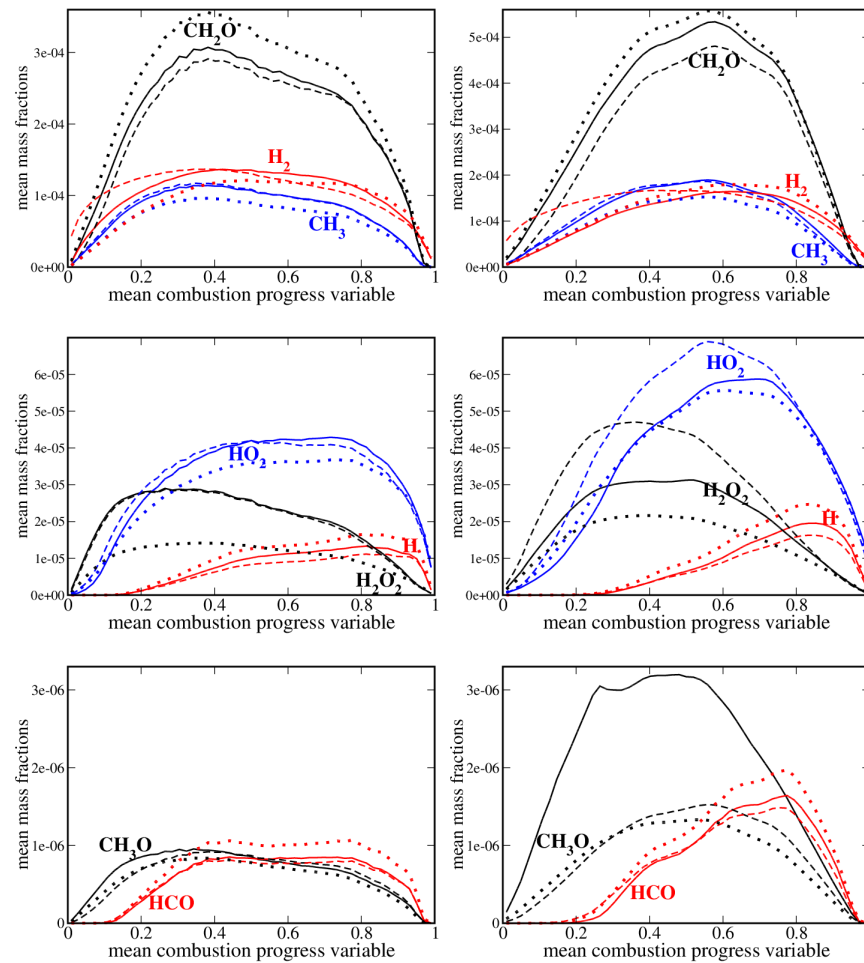
Generally speaking, since the peak value of  $|d^2Y_n/dc_k^2|$  obtained from an unperturbed laminar flame characterizes the degree of non-linearity of the dependence  $Y_{n,L}(c_k)$  in the flame, comparison of  $|d^2Y_{n,L}/dc_k^2|$  could be used for selecting the most appropriate combustion progress variable before applying Eqs. (2)-(4) to modeling premixed turbulent combustion.

Second, recent DNS data<sup>80-84</sup> indicate that the local flame structure (i) is less sensitive to preferential diffusion and Lewis number effects in more intense turbulence and (ii) tends to the structure of the equidiffusive laminar premixed flame with increasing  $Ka$  (note that burning velocity remains highly sensitive to the aforementioned effects even in very intense turbulence, as well documented in earlier experiments reviewed elsewhere<sup>85,86</sup> and in more recent measurements<sup>87-89</sup>). Accordingly, the profiles of  $Y_{n,L,Le=1}(c_T)$ , obtained from the studied unperturbed laminar premixed flame by setting Lewis numbers equal to unity for all species and reported in dashed lines in Fig. 10, were averaged adopting Eq. (2) with the actual PDF extracted from the DNS data. In Fig. 11, results obtained from flames A1 and A3 are plotted in dotted lines for some species, whereas dashed lines show  $\bar{Y}_n(\bar{c}_T)$  computed for  $Le_n \neq 1$ , with all other things being equal. In flame A1 ( $Ka = 6.0$ ), the use of the  $Y_{n,L,Le=1}(c_T)$ -profiles results in worse agreement with the DNS data (solid lines) for  $\text{CH}_2\text{O}$ ,  $\text{CH}_3$ ,  $\text{HO}_2$ ,  $\text{H}_2\text{O}_2$ ,  $\text{H}$ , and  $\text{HCO}$ , but weakly affects the agreement for  $\text{H}_2$  and  $\text{CH}_3\text{O}$ . On the contrary, the use of the  $Y_{n,L,Le=1}(c_T)$ -profiles substantially (slightly) improves predictions for  $\text{H}_2$  and  $\text{HO}_2$  ( $\text{CH}_2\text{O}$  and  $\text{H}_2\text{O}_2$ , respectively) in flame A3 ( $Ka = 540$ ). Nevertheless, even in flame A3, the use of the  $Y_{n,L,Le=1}(c_T)$ -profiles yields worse results for  $\text{CH}_3$ ,  $\text{H}$  (to a lesser extent), and  $\text{HCO}$ . For other species that are not shown in Fig. 11 differences between results simulated invoking  $Y_{n,L}(c_T)$  and  $Y_{n,L,Le=1}(c_T)$  are small. Thus, eventual mitigation of preferential diffusion effects in highly turbulent flames could explain some differences between the profiles of  $\bar{Y}_n(\bar{c}_T)$  extracted from the DNS and the profiles of  $\bar{Y}_n(\bar{c}_T)$  yielded by Eq. (2) with  $Y_{n,L}(c_T)$  and the actual PDF. However, such an explanation is not sufficient for all species, e.g.  $\text{H}$  or  $\text{CH}_3\text{O}$ . The above discussion and Fig. 11 imply that the boundary of utility of the laminar-flame profiles  $Y_{n,L}(c_T)$  for evaluating mean species concentrations in turbulent flames is close to  $Ka = O(500)$  for the studied lean methane-air mixture. Simulations at a higher  $Ka$  are required to test this hypothesis. Moreover, averaging profiles of  $Y_{n,L}(c_T)$  and  $Y_{n,L,Le=1}(c_T)$  obtained from strained laminar premixed flames could be performed to further explore physical mechanisms that reduce predictive capabilities of Eq. (2) and this could be a subject for future study.

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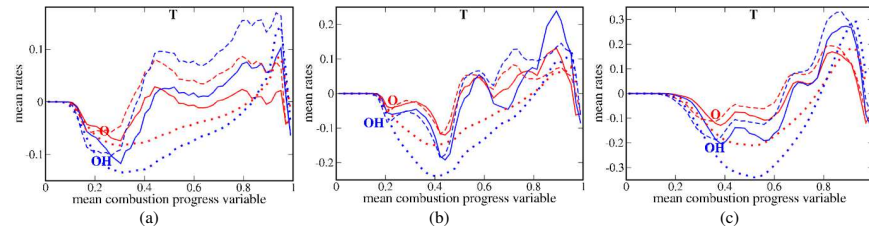
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385 The major result the present work consists in showing that Eq. (2) supplemented with the simplest version of flamelet  
386 library (a single unperturbed laminar premixed flame) works well for various intermediate species even at high  $Ka$ . This result  
387 implies also that even if strain effects play an important role locally, they significance is substantially reduced after averaging,  
388 at least for mean species concentrations. In this regard, it is worth noting that recent experimental data by Skiba et al.<sup>49</sup> show  
389 that the profiles of  $Y_n(c_T)$  calculated for freely propagating laminar premixed flame agree well with conditioned profiles of  
390  $\langle Y_n | c_T \rangle$  extracted from highly turbulent flames for  $\text{CH}_2\text{O}$ ,  $\text{CH}$ , and  $\text{OH}$ , see Figs. 2 and 3 in the cited paper.



391 **FIG. 11.** Mean mass fractions of various species noted near relevant curves vs. mean temperature-based combustion progress  
392 variable  $\bar{c}_T$ . Solid lines show  $\bar{Y}_n$  extracted from the DNS data. Dashed and dotted lines show  $\bar{Y}_n$  evaluated using the PDF  
393 extracted from the DNS data and flamelet libraries calculated for  $Le_n \neq 1$  and  $Le_n = 1$ , respectively. Results obtained from  
394 flames A1 ( $Ka = 6.0$ ) and A3 ( $Ka = 540$ ) are plotted in the left and right columns, respectively.

Contrary to Eqs. (2)-(4), results obtained by testing the flamelet Eq. (1) are less satisfactory. For instance, Fig. 12 (results computed using other  $c_k$  are worse and not reported here) show that, for major radicals such as O and OH, the profiles of  $\bar{W}_n(\bar{c}_T)$  extracted directly from the DNS data, see solid lines, differ significantly from the profiles of  $\bar{W}_n(\bar{c}_T)$  obtained by substituting the actual PDF into Eq. (1), see dashed lines. The  $\beta$ -function PDF yields even worse results, see dotted lines. The significant difference between the predictive capabilities of the flamelet Eqs. (1) and (2), cf. Fig. 12 with the next to the bottom row in Fig. 5, is associated with the fact that variations in a species concentration in a flame are smoother than variations in the rate of production/consumption of the same species. The significant differences between the mean rates  $\bar{W}_O(\bar{c}_T)$  or  $\bar{W}_{OH}(\bar{c}_T)$ , computed by adopting the actual and  $\beta$ -function PDFs, with all other things being equal, indicate limitations of the latter PDF, which were already shown in Fig. 9.



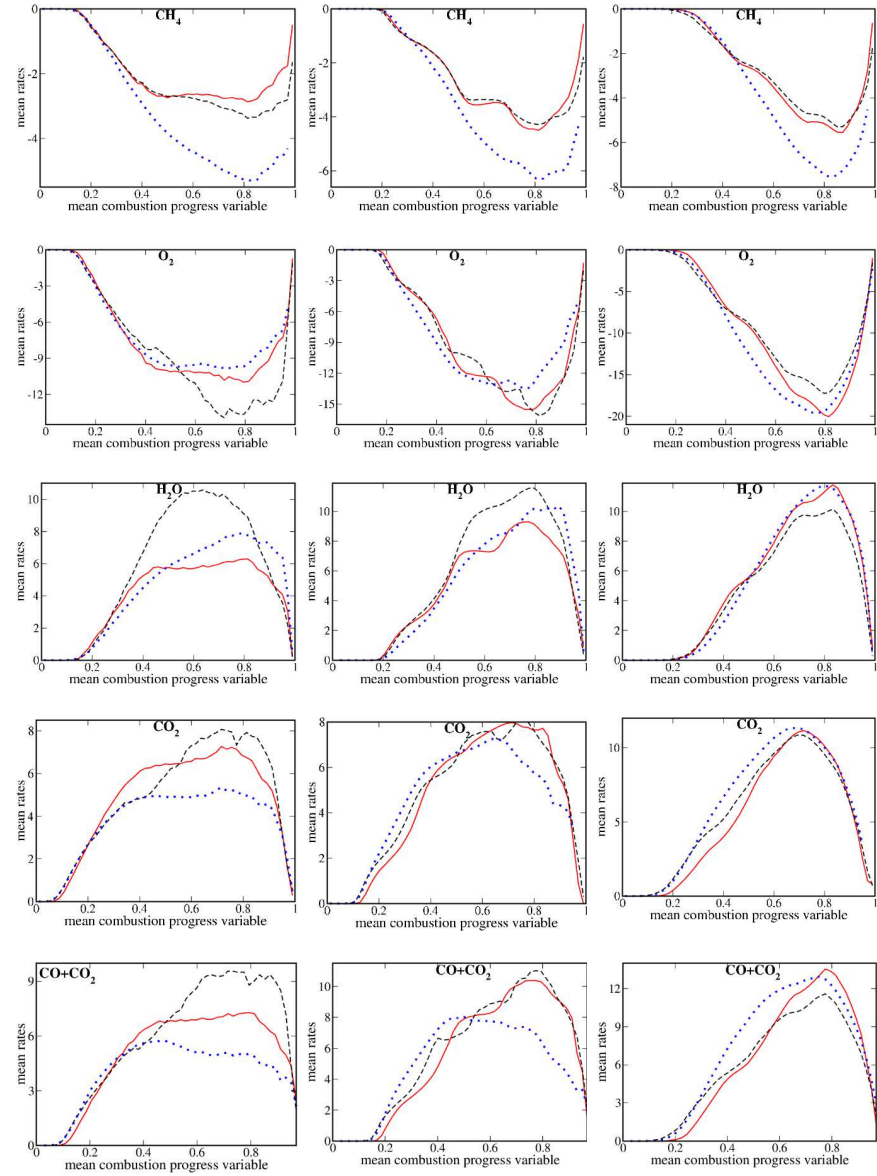
**FIG. 12.** Mean rates  $\bar{W}_n$  [ $s^{-1}$ ] of production/consumption of radicals O (red lines) and OH (blue lines) vs. mean temperature-based combustion progress variable  $\bar{c}_T$ . Solid lines show  $\bar{W}_n$  extracted from the DNS data. Dashed lines show  $\bar{W}_n$  evaluated using Eq. (1) and the PDF extracted from the DNS data. Dotted lines show  $\bar{W}_n$  calculated invoking the  $\beta$ -distribution PDF. Results computed in cases (a) A1, (b) A2, and (c) A3 are plotted in the left, middle, and right columns, respectively.

It is worth remembering, however, that Eq. (1) is commonly applied solely to evaluating the source term  $\bar{W}_c$  in the transport equation for the mean combustion progress variable,<sup>21-45</sup> whereas mean concentrations of various species are calculated using Eq. (2). Accordingly, the focus of assessment of Eq. (1) should be placed on its ability to predict  $\bar{W}_c$  for differently defined  $c_k$ . Such results are reported in Figs. 13 and 14 for species-based and temperature-based combustion progress variables, respectively. The following trends are worth noting.

First, for all  $c_k$  and in all cases, the actual and  $\beta$ -function PDFs yield different results, cf. dashed and dotted lines. Nevertheless, in most such cases, with the exception of the fuel-based  $c_1$  in all three flames,  $c_4$  in flames A1 and A2, or  $c_5$  in flames A2 and A3, either the mean rates obtained using the two PDFs show comparable agreement with the raw DNS data or the mean rates evaluated invoking the presumed  $\beta$ -function PDF agree better with the raw data, e.g.  $\bar{W}_{c,2}(\bar{c}_2)$  in flame A1,  $\bar{W}_{c,3}(\bar{c}_3)$  in all three flames, or  $\bar{W}_{c,6}(\bar{c}_T)$  in flames A2 and A3. This observation implies that, in the discussed cases, errors due to the use of the flamelet library for the reaction rates and errors due to the use of the  $\beta$ -function PDF occasionally counterbalance one another and make a wrong impression that Eqs. (1), (5), and (6) are well validated. However, “validation” of Eq. (1) by adopting a wrong PDF is definitely not validation. This example shows that *a posteriori* study performed by invoking several different submodels could lead to a wrong conclusion such as “validation” of a wrong submodel.

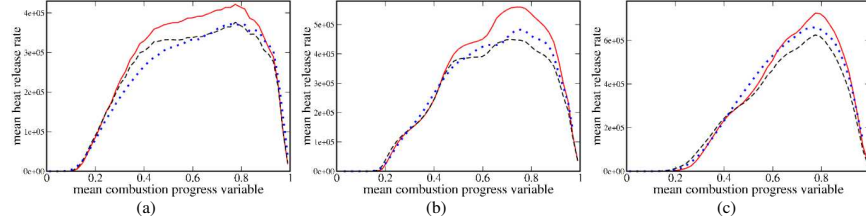
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422 **FIG. 13.** Mean rates  $\bar{W}_n$  [ $\text{s}^{-1}$ ] of production/consumption of major products/reactants vs. mean combustion progress variables  
 423 defined using the mass fraction  $Y_n$  of the same product/reactant. Legends are explained in caption to Fig. 12.





**FIG. 14.** Mean heat release rates  $\bar{W}_T$  [K/s] vs. mean temperature-based combustion progress variable  $\bar{c}_T$ . Legends are explained in caption to Fig. 12.

Second, if the cases A1, A2, and A3 are considered all together, substitution of the  $\beta$ -function PDFs given by Eqs. (5) and (6) into Eq. (1) does not allow us to predict  $\bar{W}_{c,k}$  for any  $c_k$ , cf. dotted and solid lines. In a single case, the use of certain  $c_k$  (e.g.,  $c_2$  in case A1,  $c_2$  or  $c_3$  in case A2,  $c_3$  or  $c_T$  in case A3) can yield good results due to the aforementioned mutual cancellations of two types of errors. Indeed, in each of these five cases, the use of the actual PDF yields the profile of  $\bar{W}_{c,k}(\bar{c}_k)$  that differs substantially from  $\bar{W}_{c,k}(\bar{c}_k)$  calculated by adopting a less accurate  $\beta$ -function PDF.

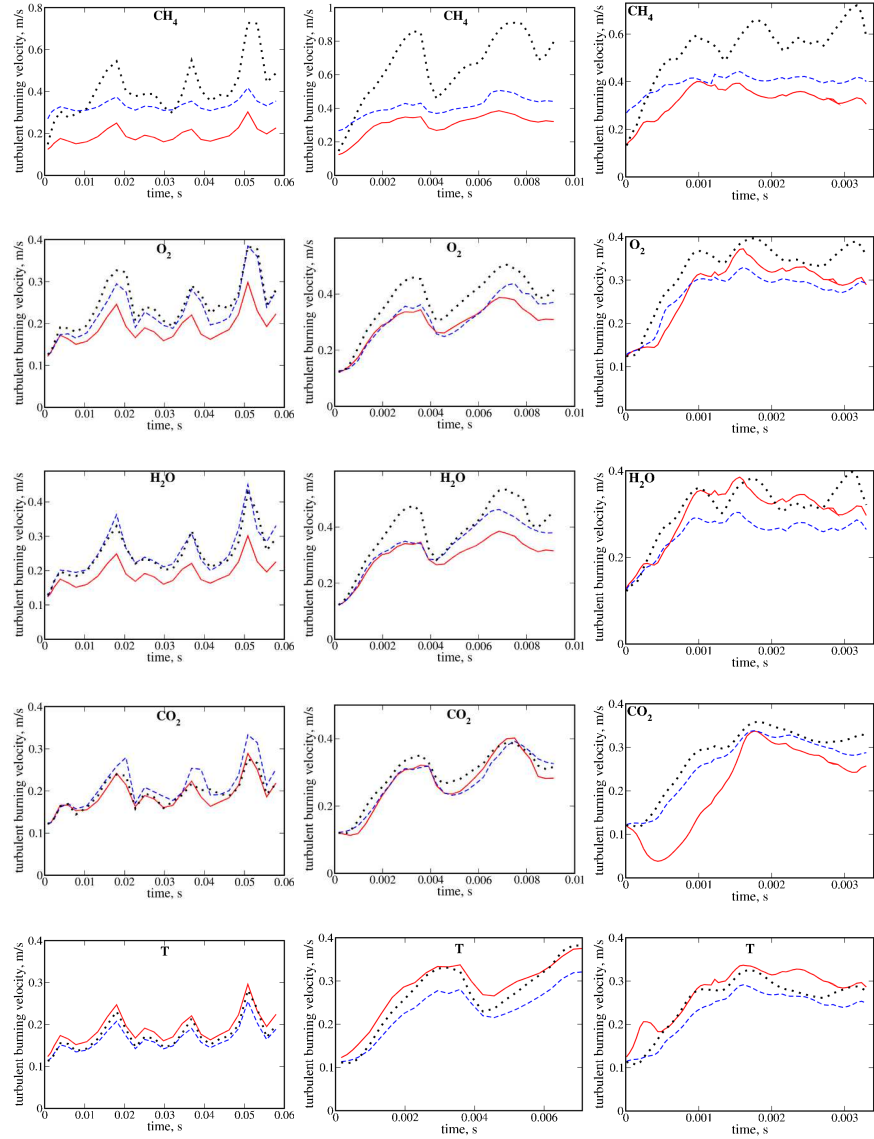
Third, dependencies of  $\bar{W}_{c,k}(\bar{c}_k)$  calculated by substituting the actual PDF into Eq. (1) differ substantially from dependencies of  $\bar{W}_{c,k}(\bar{c}_k)$  extracted directly from the DNS data for  $\bar{c}_2$  (with the exception of case A2),  $\bar{c}_3$ ,  $\bar{c}_4$  or  $\bar{c}_5$  (in case A1), and  $\bar{c}_T$ . The differences (i) are less pronounced in case A3 and (ii) are small for the fuel-based  $\bar{c}_1$  (with the exception of the trailing edges of the flame brushes in cases A1 and A2). Therefore, as far as modeling of the source term  $\bar{W}_c$  in the transport equation for the mean combustion progress variable is concerned, Figs. 13 and 14 highlight the fuel-based  $c_1$  and, to a lesser extent, the  $\text{CO}_2$ -based  $c_4$ . On the contrary,  $c_1$  is not the best choice for evaluating the mean mass fraction of CO using Eq. (2), cf. red solid and dashed lines in the top row in Fig. 4, or the mean temperature using Eq. (3), cf. solid and dashed lines in the top row in Fig. 1. The  $\text{CO}_2$ -based  $c_4$  or  $c_5$  is the worst choice for calculating the mean temperature and density adopting Eqs. (3) and (4), respectively, cf. solid and dashed lines in the fourth rows in Figs. 1 and 2, respectively.

It is of interest to note that turbulent burning velocities obtained by integrating different  $\langle W \rangle_{c,k}[\langle c \rangle_k(x, t)]$  along the normal to the mean flame brush can be substantially different even if the actual  $\bar{W}_{c,k}(\bar{c}_k)$  appears to be close to  $\bar{W}_{c,k}(\bar{c}_k)$  evaluated by substituting the actual PDF into Eq. (1). It is worth remembering that  $\langle W \rangle_{c,k} = \langle W \rangle_k[\langle c \rangle_k(x, t)]$  and  $\bar{W}_{c,k}[\bar{c}_k(x)]$  designate the rates averaged over the transverse plane at a single instant and the rates averaged over the transverse plane and various instants, respectively.

The aforementioned difference is reported in Fig. 15, which shows evolutions of turbulent burning velocities defined as follows

$$U_{T,k}(t) = \frac{1}{\rho_u(Y_{k,b} - Y_{k,u})} \int_{-\infty}^{\infty} \langle \rho \rangle(x, t) \langle W \rangle_k[\langle c \rangle_k(x, t)] dx \quad (7)$$

for species-based combustion progress variables  $c_1$  ( $\text{CH}_4$ ),  $c_2$  ( $\text{O}_2$ ),  $c_3$  ( $\text{H}_2\text{O}$ ), and  $c_4$  ( $\text{CO}_2$ ) or



448 **FIG. 15.** Evolution of turbulent burning velocities evaluated using different combustion progress variables specified in the top  
449 of each subfigure. Solid lines show  $U_{T,k}(t)$  calculated adopting  $\bar{W}_k$  extracted from the DNS data. Dashed lines show  $U_{T,k}(t)$   
450 obtained using Eqs. (1), (7) or (8) and the PDF extracted from the DNS data. Dotted lines show  $U_{T,k}(t)$  computed invoking the  
451  $\beta$ -distribution PDF. Results obtained in cases A1, A2, and A3 are plotted in the left, middle, and right columns, respectively.

$$U_{T,6}(t) = \frac{1}{\rho_u(T_b - T_u)} \int_{-\infty}^{\infty} \langle \rho \rangle(x, t) \langle W \rangle_6[\langle c \rangle_6(x, t)] dx \quad (8)$$

for the temperature-based combustion progress variable  $c_6$ . For the fuel-based  $c_1$ , the actual  $U_{T,1}(t)$ , see solid lines in the top row, is substantially lower than  $U_{T,1}(t)$  yielded by Eq. (1) with the actual PDF, see dashed lines, whereas the corresponding dependencies of  $\bar{W}_{c,1}(\bar{c}_1)$  appear to be close to one another in the largest parts of the mean flame brushes, cf. solid and dashed lines in the top row in Fig. 13. This apparent inconsistency is associated with substantial contribution to the integral in Eq. (7) from thick zones characterized by large  $\bar{c}_1$ , where the two  $\bar{W}_{c,1}(\bar{c}_1)$  differ from one another and the spatial gradient  $d\bar{c}_1/dx$  is relatively low. Therefore, while results plotted in Figs. 13 and 14 highlight the fuel-based  $c_1$ , Fig. 15 does not do so.

On the contrary, Fig. 15 shows that, among the investigated  $c_k$ , the best agreement between the actual  $U_{T,k}(t)$  and  $U_{T,k}(t)$  yielded by Eq. (1) with the actual PDF has been obtained for the temperature-based  $c_6$  and CO<sub>2</sub>-based  $c_4$  in case A1, the CO<sub>2</sub>-based  $c_4$  and O<sub>2</sub>-based  $c_2$  in case A2, and the O<sub>2</sub>-based  $c_2$ , temperature-based  $c_5$ , and fuel-based  $c_1$  in case A3. However, for instance, the dependencies of  $\bar{W}_{c,4}(\bar{c}_4)$ , plotted in solid and dashed lines in the left column in the fourth row in Fig. 13, are substantially different in case A1. Accordingly, comparison of Figs. 13 and 15 implies that the good results reported for the CO<sub>2</sub>-based  $c_4$  in case A1 in the latter figure stem, at least in part, from occasional mutual cancellation of errors in evaluation of  $\bar{W}_{c,4}(\bar{c}_4)$  in different zones of the mean flame brush. This example demonstrates again the importance of using several different tests in a validation study.

If all three cases are considered together, Fig. 15 highlights the O<sub>2</sub>-based  $c_2$ , the CO<sub>2</sub>-based  $c_4$ , and, to a lesser extent, the temperature-based  $c_6$ . In particular, Eqs. (7) and (1) with the actual PDF perform excellent for the CO<sub>2</sub>-based  $c_4$  in cases A1 and A2 but fail in case A3. Moreover, the use of  $c_4$  does not allow Eqs. (3) and (4) to predict the mean temperature and density, respectively, see the fourth rows in Figs. 1 and 2, respectively. Furthermore, Eq. (2) with  $c_4$  performs substantially worse for CH<sub>4</sub>, CO, see the fourth row in Fig. 4, and many intermediate species when compared to the same Eq. (2) with the temperature-based  $c_6$ . As far as the O<sub>2</sub>-based  $c_2$  is concerned, its use yields worse results for the mean density, see Fig. 1, as well as mass fractions of CO, see Fig. 4, CH<sub>2</sub>O, see Fig. 5, CH<sub>3</sub>, see Fig. 6, HCO and CH<sub>3</sub>O, see Fig. 8.

#### IV. IMPLICATIONS FOR MODELING

The present DNS data show that Eq. (1), Eqs. (2)-(4), and Eq. (7) or (8) perform differently for differently defined combustion progress variables. In particular, Eqs. (2)-(4) perform best for the temperature-based  $c_6$ . However, application of Eq. (1) and Eq. (8) to  $c_6$  yields underestimated  $\bar{W}_{c,6}(\bar{c}_6)$  and  $U_{T,6}(t)$ . Equation (1) performs best for the fuel-based  $c_1$  and, to a lesser extent, for the CO<sub>2</sub>-based  $c_4$ , whereas Eq. (7) performs best for the O<sub>2</sub>-based  $c_2$  and for the CO<sub>2</sub>-based  $c_4$ . However, as discussed earlier, Eqs. (2)-(4) perform substantially worse with  $c_1$ ,  $c_2$ ,  $c_4$  or  $c_5$  when compared to  $c_T$ . Thus, the present results considered all together imply that mean mass fractions of various species can be evaluated by adapting Eq. (2) independently of Eq. (1), for example, by invoking a model of the mean (or filtered) rate  $\bar{W}_c$ , which performs better than Eq. (1). The reader interested in such models is referred to review literature.<sup>86,90-94</sup>

482 If an appropriate model of the influence of turbulence on premixed combustion is invoked and the mean fields of  $\bar{c}$  and  
483  $\bar{W}_c$  are obtained either directly within the RANS framework or by averaging the counterpart filtered fields within the LES  
484 framework, the mean mass fractions of various species can simply be calculated at a post-processing stage of the simulations  
485 using Eq. (2). To do so, not only a closure relation for the mean (or filtered) rate  $\bar{W}_c$ , but also a PDF  $P(c, \mathbf{x}, t)$  are required and  
486 modeling the PDF still challenges the combustion community. The issue could be addressed by developing the approach that  
487 deals with a transport equation for the PDF.<sup>4,11-14</sup> This research direction appears to be prioritized from the fundamental  
488 perspective and the present study provides additional motivation for developing it.

489 Nevertheless, from the application perspective, the presumed PDF approach may also deserve development, e.g. by taking  
490 the following opportunity. If Eq. (1) is not applied to close the mean rate  $\bar{W}_c$ , but another model of  $\bar{W}_c$  is invoked, then the  
491 following equation

$$\bar{c}^2(\mathbf{x}, t) = \int_0^1 c^2 P(c, \mathbf{x}, t) dc, \quad (9)$$

492 which is commonly used to evaluate unknown parameters of a presumed PDF  $P(c, \mathbf{x}, t)$ , could be substituted with a constraint  
493 of

$$\bar{W}_c(\mathbf{x}, t) = \int_0^1 W_c(c) P(c, \mathbf{x}, t) dc. \quad (10)$$

494 Therefore, the presumed PDF is calibrated by (i) invoking a closure relation for  $\bar{W}_c(\mathbf{x}, t)$  yielded by another model that is not  
495 based on a PDF  $P(c, \mathbf{x}, t)$  and (ii) adopting Eq. (10) instead of Eq. (9). The use of Eq. (10) for PDF calibration will, in particular,  
496 offer an opportunity to obtain a PDF that better predicts the probability of finding reaction zones. Indeed, the mean rate  $\bar{W}_c(\mathbf{x}, t)$   
497 is directly linked with that probability, whereas such a link appears to be doubtful for the variance  $\bar{c}^{T/2}(\mathbf{x}, t)$  or  $\bar{c}^{T/2}(\mathbf{x}, t)$ .<sup>17,86</sup>  
498 For instance, these variances are solely controlled by the probabilities of finding unburned (fresh) reactants and fully burned  
499 products in the BML limit.<sup>69</sup>

500 Encouraging results obtained in the present study by testing the flamelet Eqs. (2)-(4) suggest that the presumed PDF  
501 approach could substantially be advanced (i) adapting the classical flamelet PDF,<sup>66</sup> i.e.  $1/(\delta_L |\nabla c|_L)$ , but also (ii) invoking Eq.  
502 (10) to calibrate the PDF, as argued above. Recently, this proposal was developed<sup>51,53</sup> by analyzing DNS data obtained by Dave  
503 and Chaudhuri<sup>54</sup> and Im et al.<sup>55-59</sup> from a lean hydrogen-air flame characterized by two different equivalence ratios and four  
504 different Karlovitz numbers ranging from 0.75 to 126. Further development and assessment of such a presumed flamelet-based  
505 PDF will be a subject for future analysis of the present DNS data.

## 506 V. CONCLUDING REMARKS

507 A quantitative *a priori* assessment of the simplest version of flamelet approach to evaluating the mean density  $\bar{\rho}$ , the mean  
508 temperature  $\bar{T}$ , the mean mass fractions  $\bar{Y}_n$  of various species, and the mean rates of the species production/consumption in a  
509 premixed turbulent flame has been performed by analysing complex-chemistry DNS data obtained earlier<sup>45,60</sup> from three lean

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methane-air flames characterized by three different Karlovitz numbers ranging from 6 to 540. The approach consists in (i) simulating the unperturbed laminar flame in order to obtain dependencies of the temperature, density, and mass fractions of various species on a single combustion progress variable  $c$  and (ii) averaging these dependencies by invoking a PDF for the same combustion progress variable, see Eqs. (2)-(4). When assessing the approach, six different choices of  $c$  have been probed and the PDF (i) either has been extracted directly from the DNS data or (ii) has been modelled invoking the well-known presumed  $\beta$ -function and using the first two moments of the  $c$ -field yielded by the DNS data. A similar method, see Eq. (1), has also been applied to assessing capabilities of the flamelet approach for predicting the mean source term  $\bar{W}_c$  in the transport equation for the mean combustion progress variable.

The major results of this analysis are as follows.

- First, at all three  $Ka$ , substitution of (i) the actual PDF extracted from the DNS data and (ii) the simplest flamelet library  $\rho_L(c)$ ,  $T_L(c)$ , and  $Y_{n,L}(c)$ , computed for a single unperturbed laminar premixed flame, into Eqs. (2)-(4) has allowed us to quantitatively predict the profiles of  $\bar{\rho}(\bar{c})$ ,  $\bar{T}(\bar{c})$ , and  $\bar{Y}_n(\bar{c})$  for  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_2\text{O}$ ,  $\text{CH}_3$ , and  $\text{HCO}$  provided that the combustion progress variable is appropriately defined (it is based on the temperature for the studied lean methane-air flames). For the other seven species, with the exception of  $\text{CH}_3\text{O}$  at the highest  $Ka$ , the results are also encouraging.
- Second, the  $\beta$ -function PDF differs significantly from the actual PDF extracted from the DNS data and the use of the  $\beta$ -function PDF yields substantially worse results for intermediate species such as  $\text{OH}$ ,  $\text{O}$ ,  $\text{H}$ ,  $\text{CH}_3$ , and  $\text{HCO}$ .
- Third, for all investigated combustion progress variables, the mean rates  $\bar{W}_n$  of production/consumption of various species (e.g., the radicals  $\text{O}$  and  $\text{OH}$ ) are poorly predicted by Eq. (1) even if the actual PDF is adopted. Moreover, if all three flames are considered together, Eq. (1) does not simultaneously predict (i) the profiles  $\bar{W}_{c,k}(\bar{c}_k)$  of the mean rate of product creation and (ii) turbulent burning velocities  $U_{t,k}$  obtained by integrating these profiles. For instance,  $\bar{W}_{c,k}(\bar{c}_k)$  is reasonably well predicted adopting the fuel-based combustion progress variable but  $U_{t,k}$  is reasonably well predicted using the oxygen-based combustion progress variable.

These three major findings are consistent with recent results<sup>50,52</sup> computed by analyzing other DNS data obtained from four lean hydrogen-air premixed turbulent flames characterized by two different equivalence ratios and Karlovitz numbers ranging from 0.75 to 126. However, the present study addresses a significantly higher  $Ka = 540$  (case A3) and a more complicated chemical system (when compared to hydrogen, combustion of methane involves more reactions and more species). For instance, we are not aware of a study that shows capability of Eq. (2) for quantitatively predicting mean mass fractions of carbon-containing intermediate species (including  $\text{CO}$ ) in premixed or stratified turbulent flames. Moreover, results obtained from flame A3 indicate, for the first time to the best of the present authors knowledge, that performance of Eq. (2) in highly turbulent flames can be improved by using a flamelet library obtained from equidiffusive unperturbed laminar flame.

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Consistency of the present and recent<sup>50,52</sup> results implies that the three major findings highlighted above are sufficiently general, while other details (e.g. the best choice of a combustion progress variable) could be mixture sensitive. For instance, the fuel-based combustion progress variable performs better in the aforementioned hydrogen flames, while the temperature-based  $c$  performs sufficiently well also.

The highlighted findings imply that, in order to evaluate the mean temperature, density and species mass fractions, Eqs. (2)-(4) could be coupled with another model of premixed turbulent combustion whose predictive capabilities are better documented when compared to Eq. (1). In such a case, Eqs. (2)-(4) could be implemented as post-processing of a mean  $\bar{c}$ -field computed by numerically integrating a single transport equation for the mean combustion progress variable.

As already mentioned, the best predictions of the mean concentrations of various species in the studied lean methane-air flame were obtained using the temperature-based combustion progress variable, while this conclusion could be mixture sensitive. Selection of a progress variable  $c_k$  that yields the lowest maximum absolute value of the second derivative  $d^2Y_n/dc_k^2$  in the laminar flame and, therefore, is associated with a less non-linear profile  $Y_n(c_k)$  could be recommended for a study aimed at predicting the mean mass fraction  $\bar{Y}_n$  using Eq. (2).

It is also worth noting that the use of the profiles  $Y_{n,L,L=1}(c)$  obtained from the equidiffusive laminar flame improves predictions of mean concentrations of certain (but not all) species at the highest  $Ka$ . This observation implies that (i) the boundary of validity of Eq. (2) with the canonical laminar-flame profiles  $Y_{n,L}(c)$  is close to  $Ka = 0(540)$  for the studied lean methane-air mixture and (ii) the averaged influence of preferential diffusion phenomena on the local flame structure is reduced at higher Karlovitz numbers, in line with earlier studies.<sup>80-84</sup> If small-scale turbulent mixing changes the local flame structure, larger-scale eddies can still strain the flame. Accordingly, substitution into Eq. (2) of the profiles  $Y_{n,L,L=1}(c)$  obtained from equidiffusive strained laminar flames could be an interesting task for future research.

Similar to the vast majority of recent complex-chemistry DNS studies of highly turbulent premixed flames,<sup>55-61,78-84</sup> the present analysis is restricted to small-scale turbulence, because 3D complex-chemistry DNS of combustion in intense and large-scale turbulence is not yet computationally affordable. Accordingly, Damköhler numbers addressed in the present and other DNS works may appear to be too low when compared to conditions reached in contemporary engines. However, it is worth remembering that the explored flamelet approach is commonly considered to work better at higher Damköhler and lower Karlovitz numbers. Therefore, the conditions of the present study appear to be more challenging for its goals when compared to flames characterized by  $Da > 1$ . Accordingly, the major conclusions are expected to hold under higher Damköhler numbers.

All in all, the flamelet-based Eqs. (2)-(4) appear to be a useful CFD tool even at Karlovitz number as large as 540 provided that an appropriate definition of combustion progress variable is adopted, and the PDF is well modeled. Therefore, these results (i) indicate that the domain of the flamelet concept validity is substantially wider than it was earlier assumed, in line with recent studies,<sup>48,49</sup> and (ii) motivate modeling the PDF in Eqs. (2)-(4). Extension of the present work to filtered scalar fields and filtered density functions computed in a LES is another subject for future studies.

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## 580 DATA AVAILABILITY

581 The data that support the findings of this study are available from the corresponding author upon reasonable request.

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619 of a combustion progress variable  $c$ , but also of a mixture fraction in the case of stratified combustion, the mixture enthalpy in  
620 the case of non-adiabatic flame, pressure in the case of non-isobaric burning (e.g. in a piston engine), stretch rate in highly  
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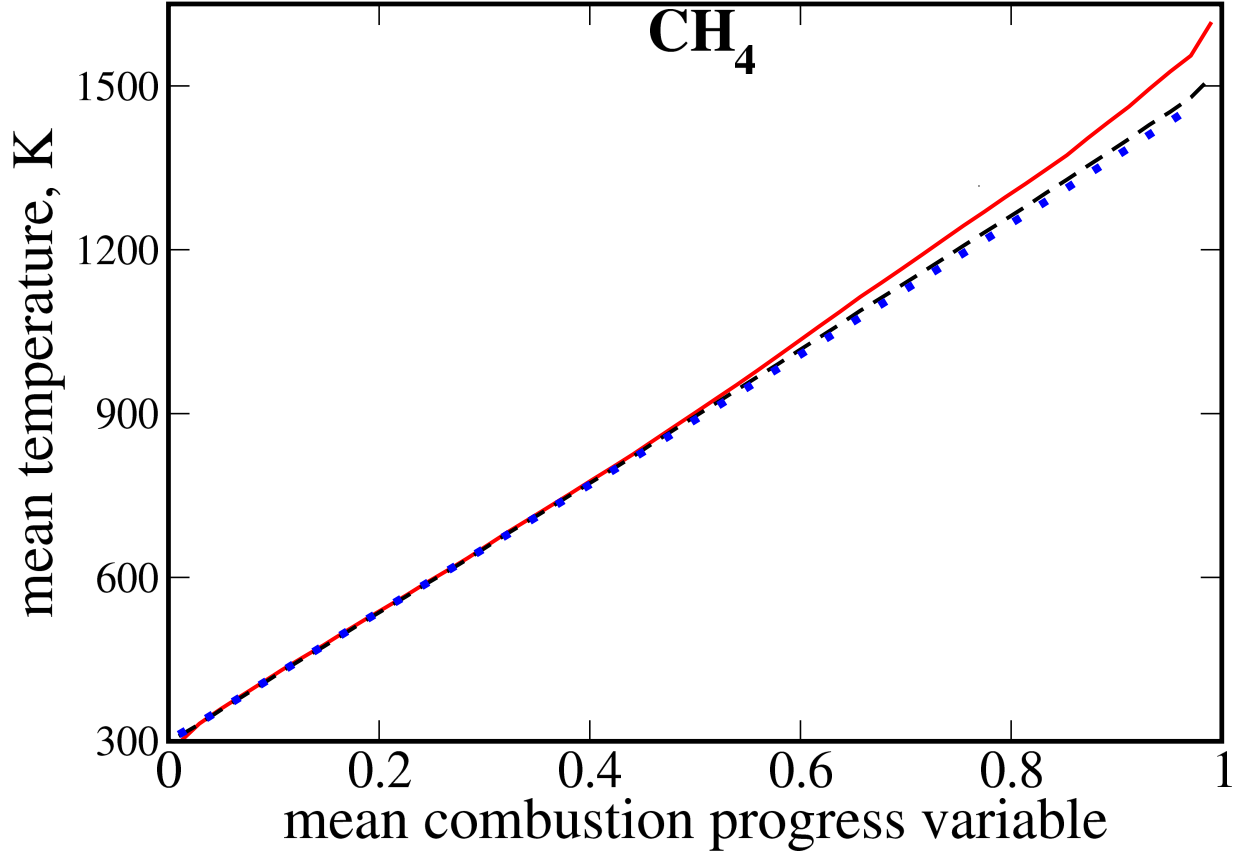
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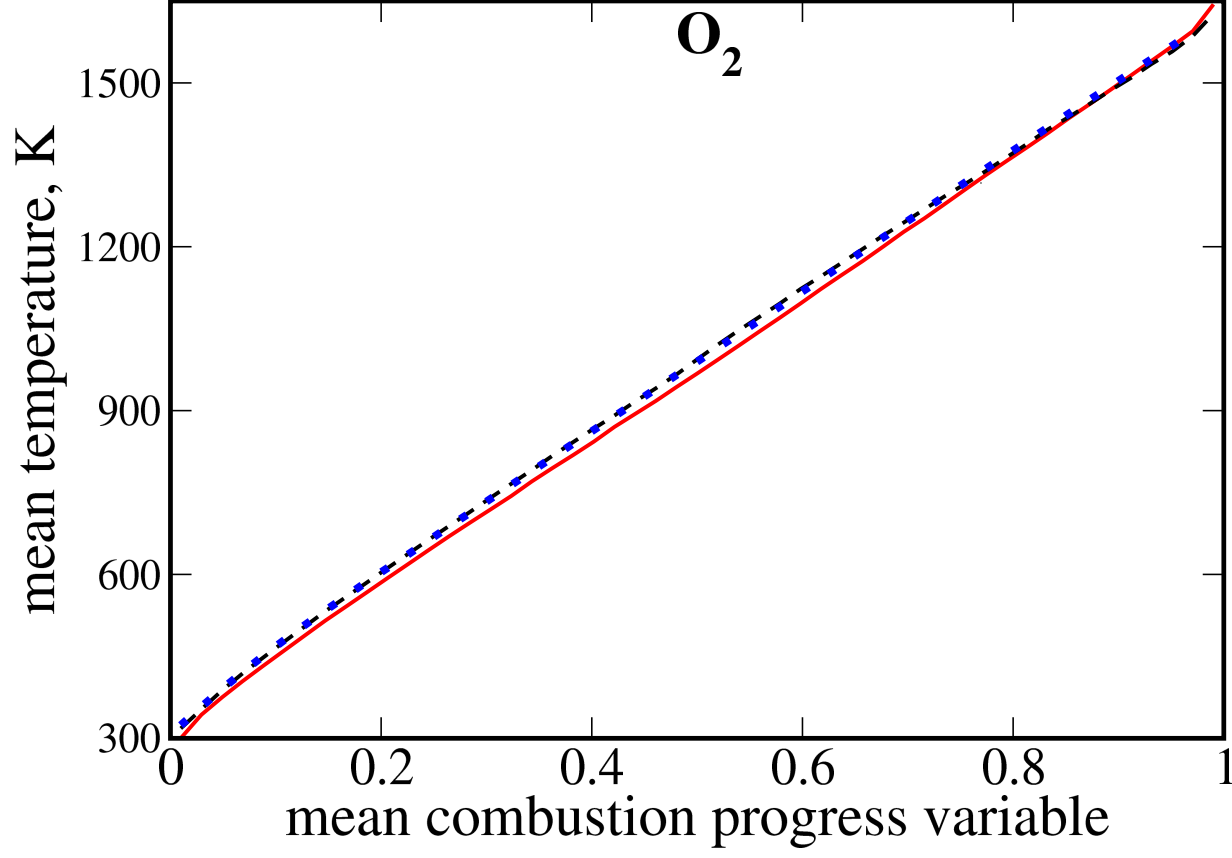
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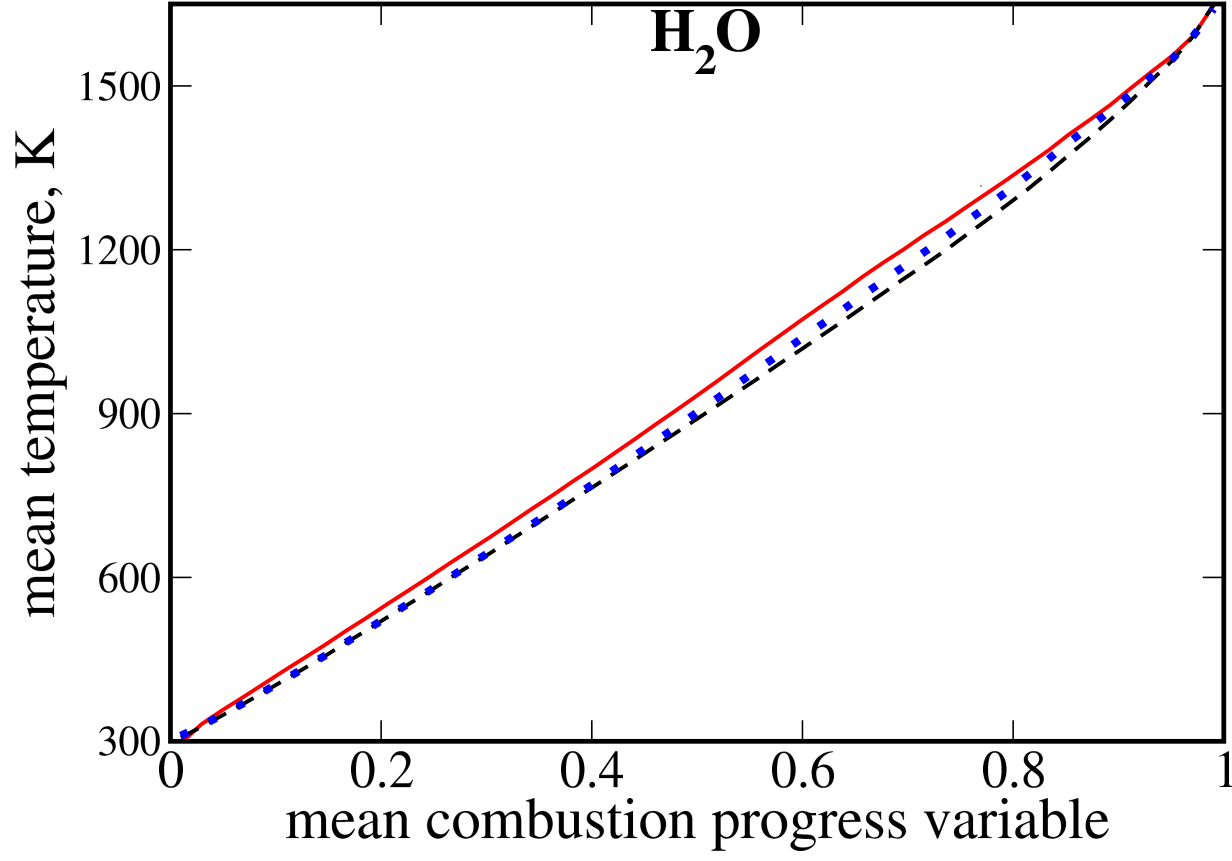
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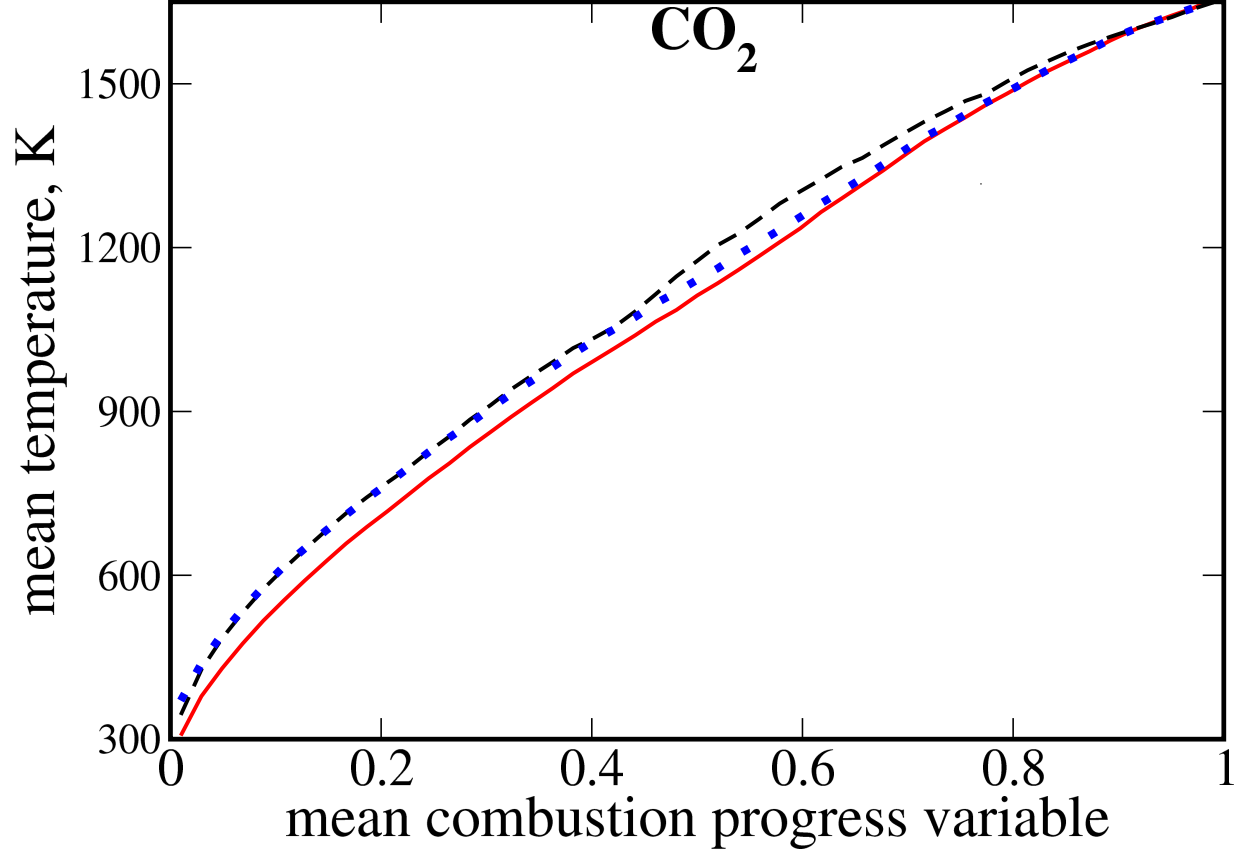
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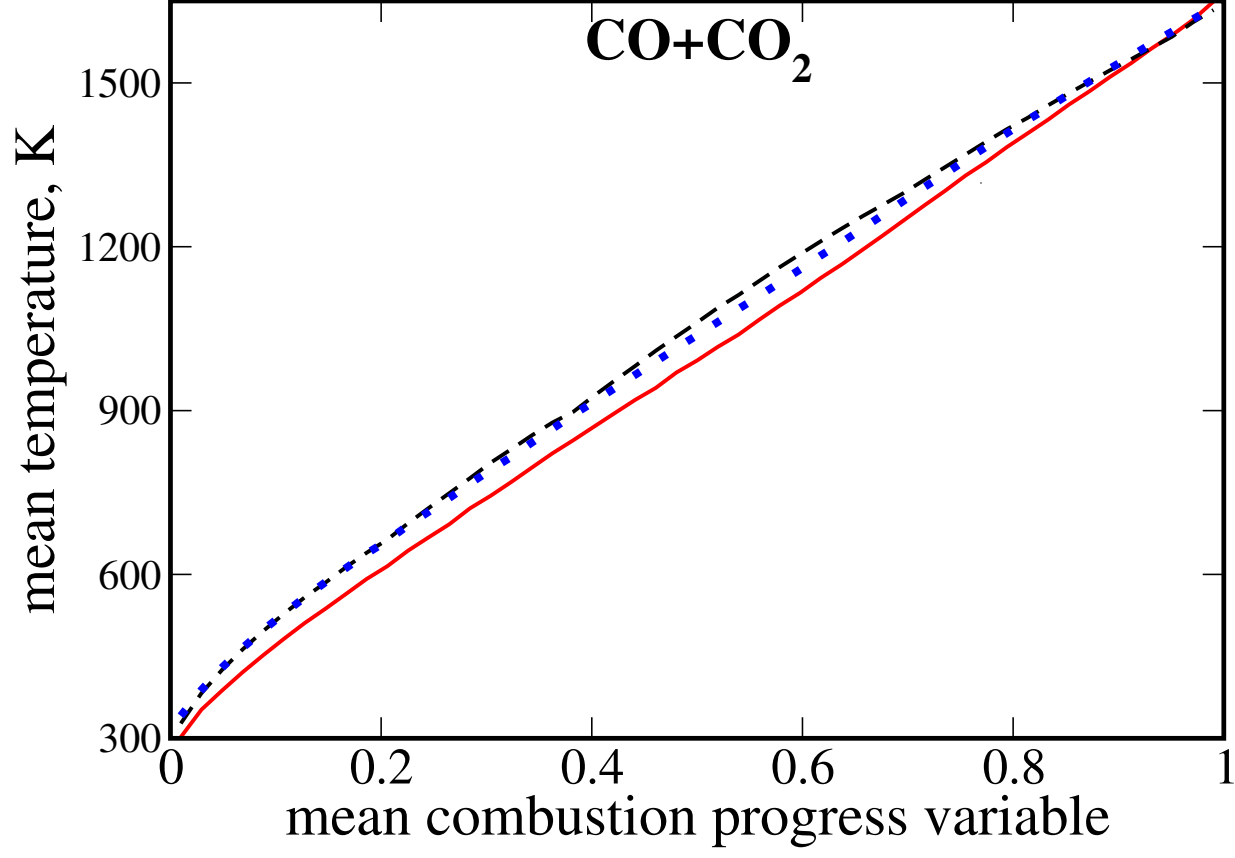
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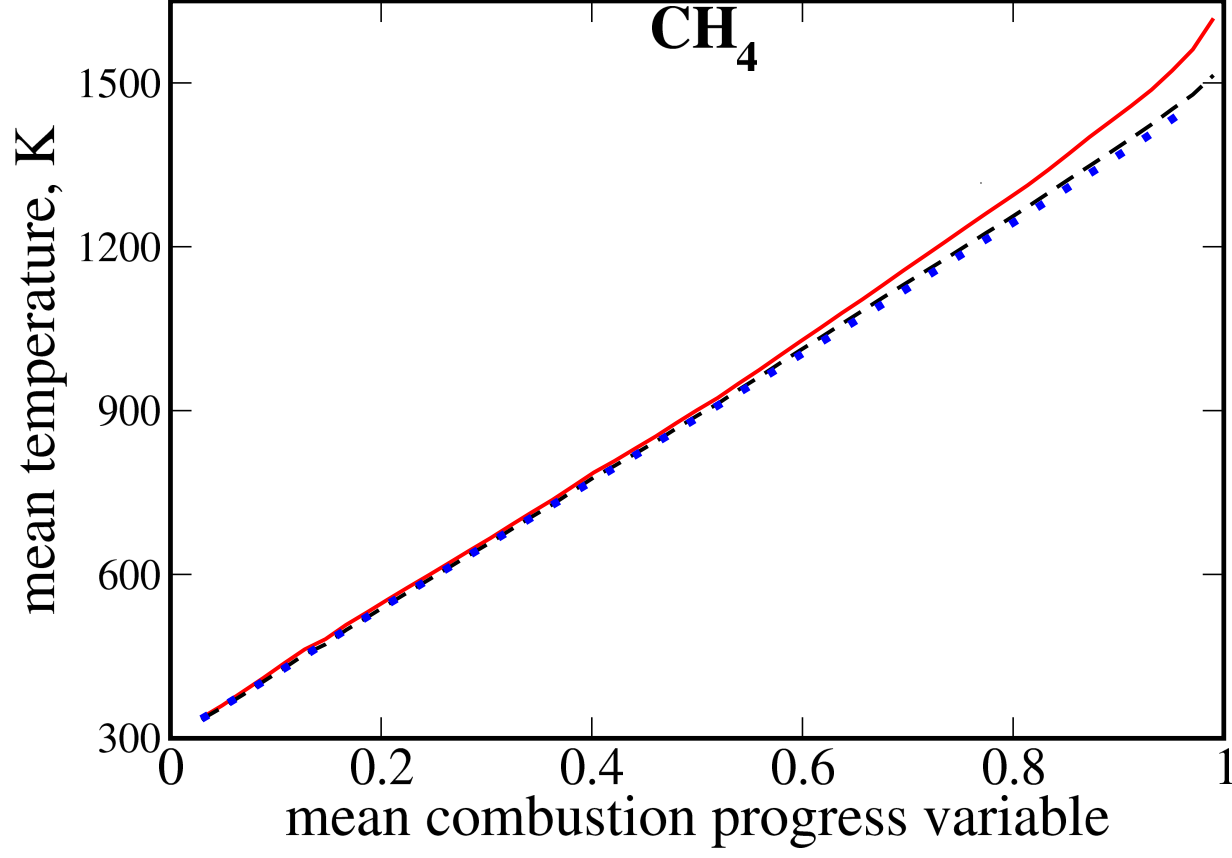
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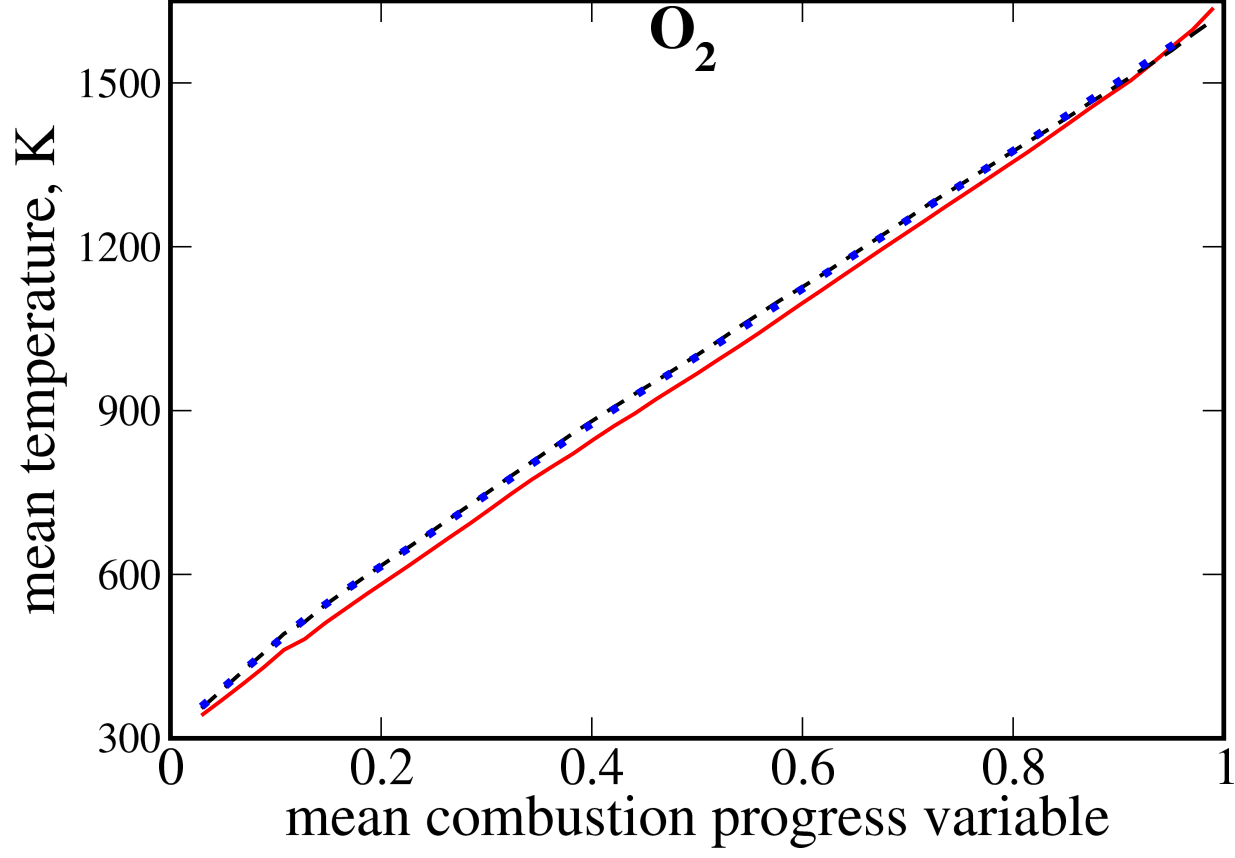
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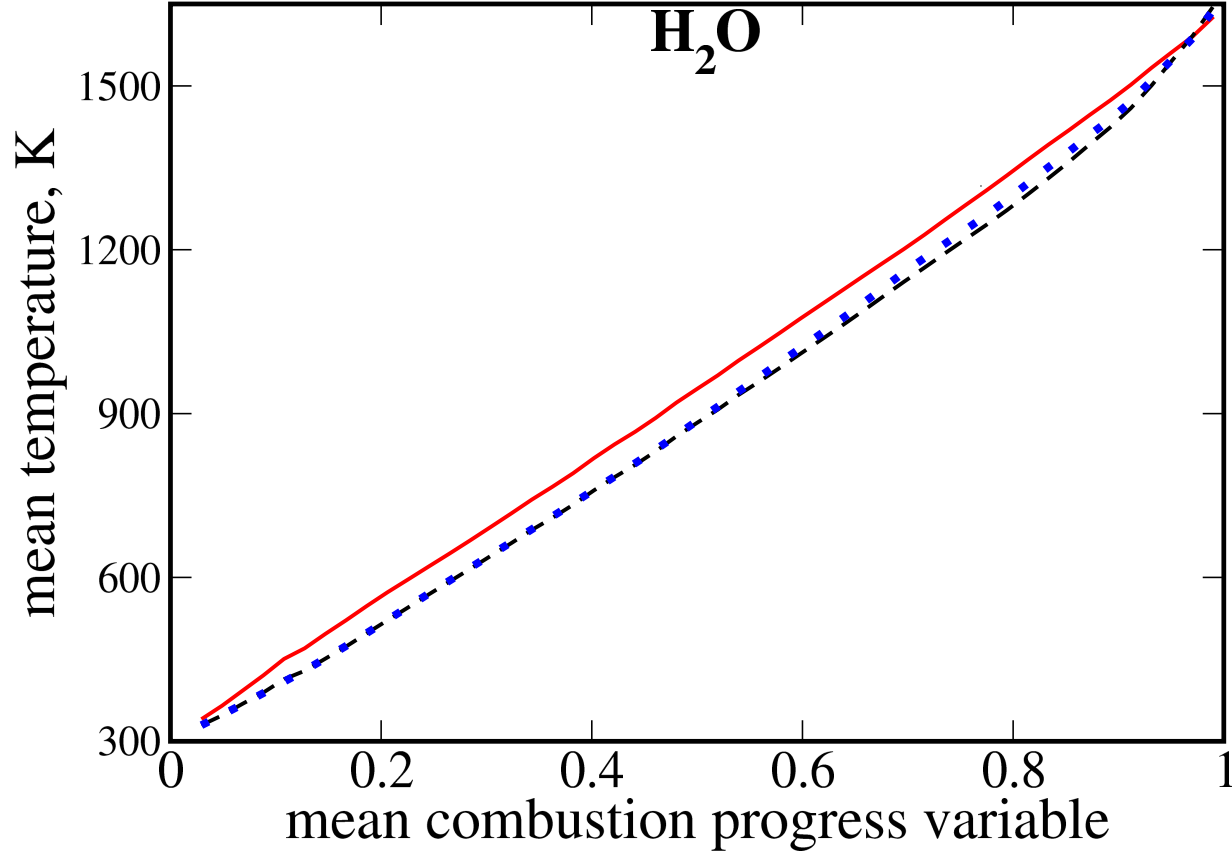
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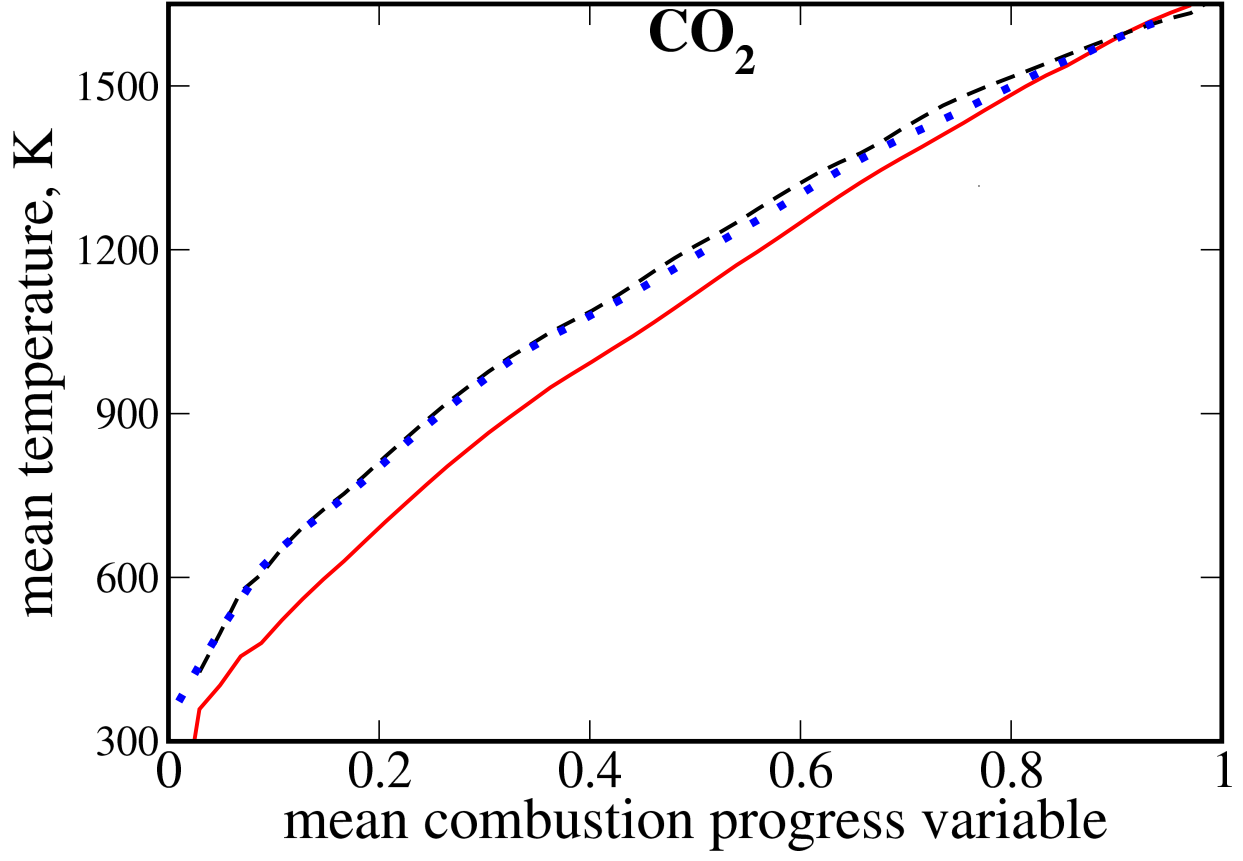
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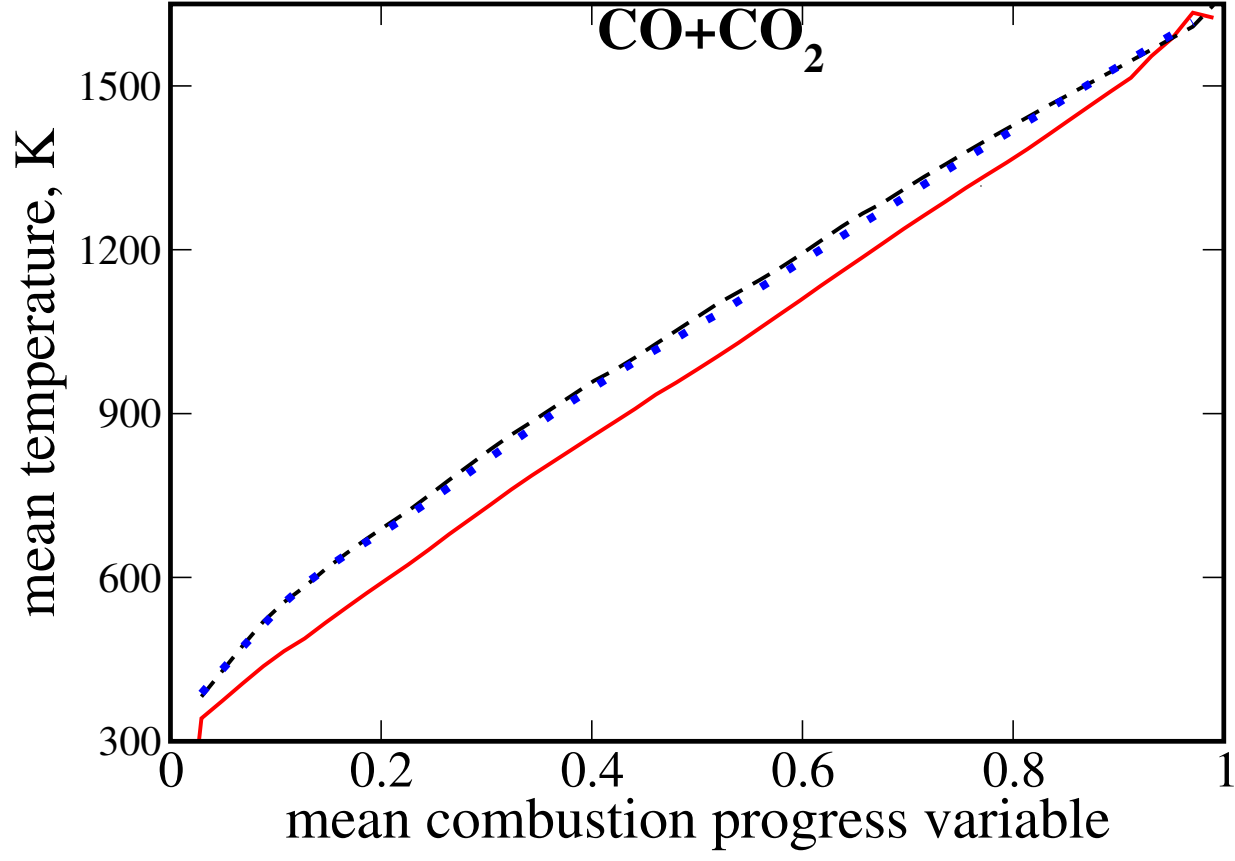
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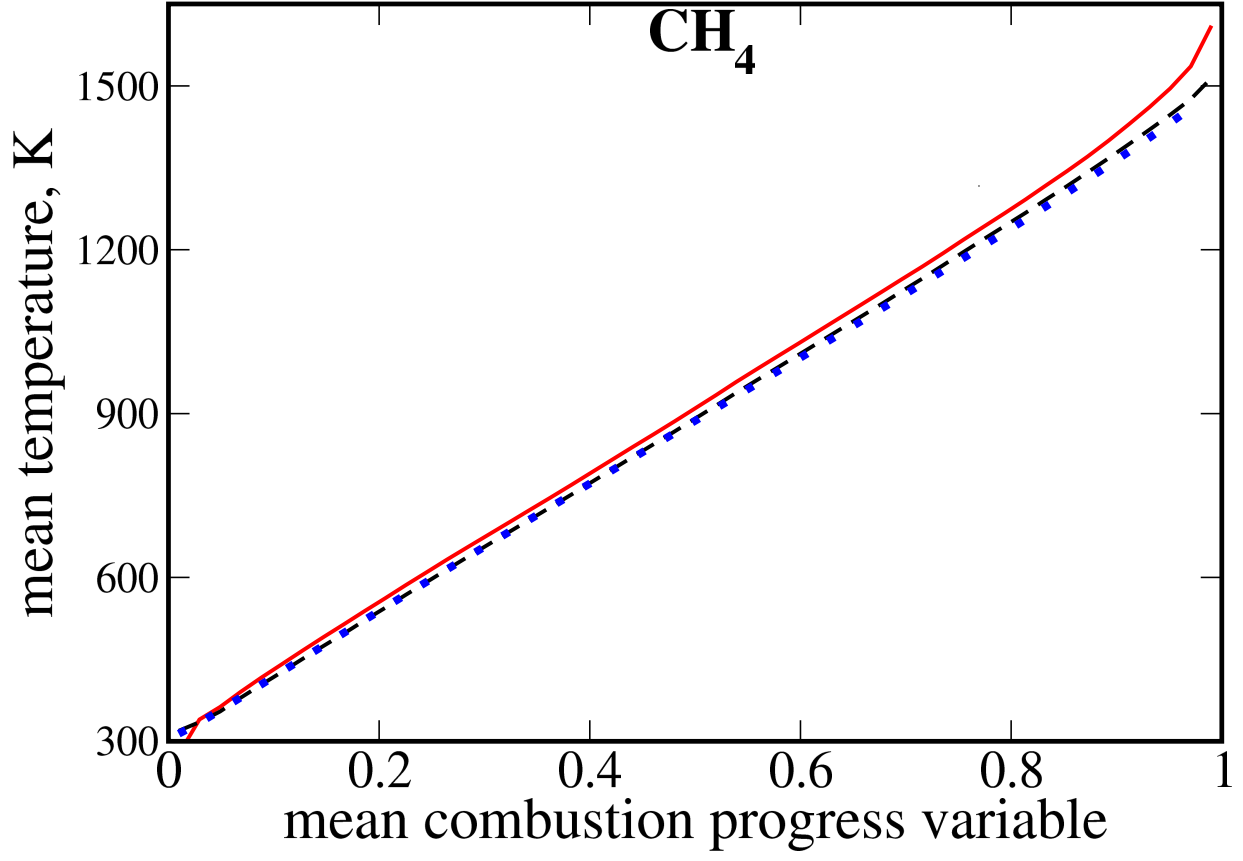
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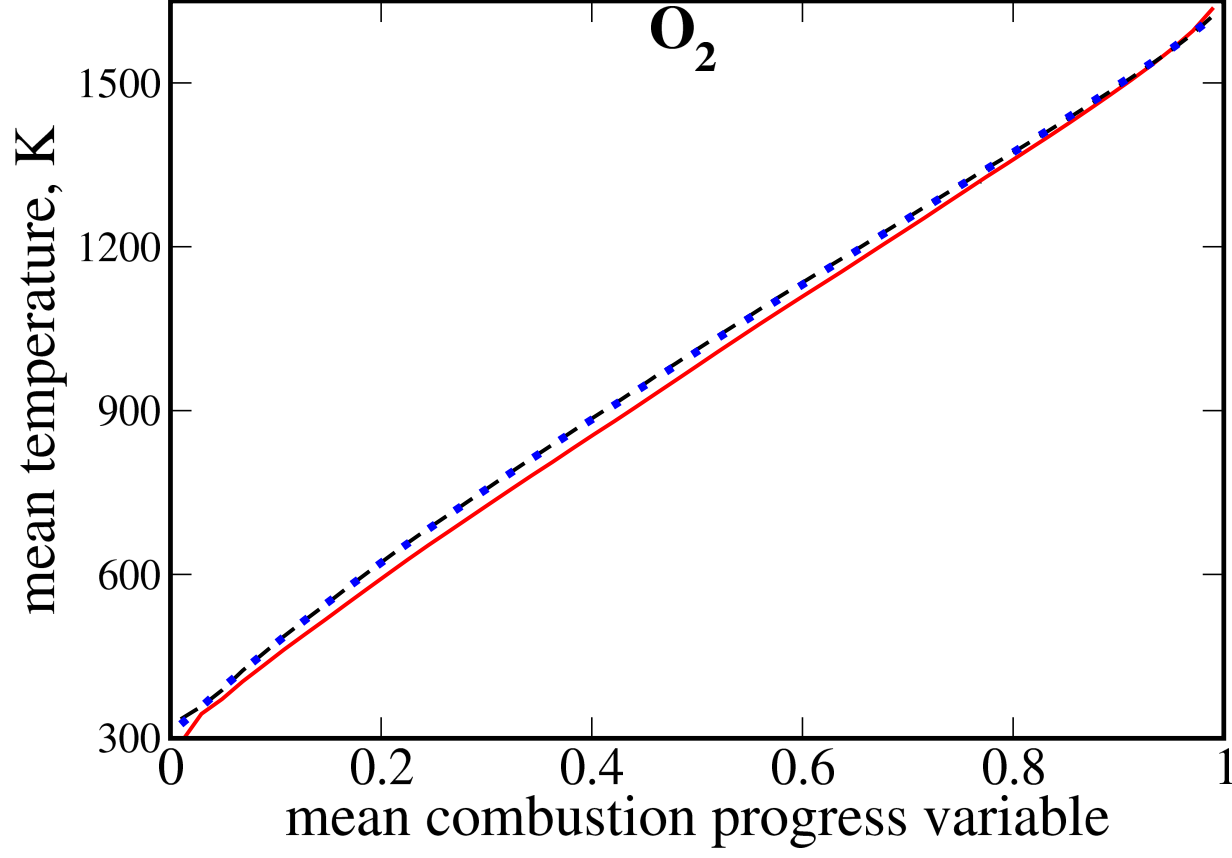
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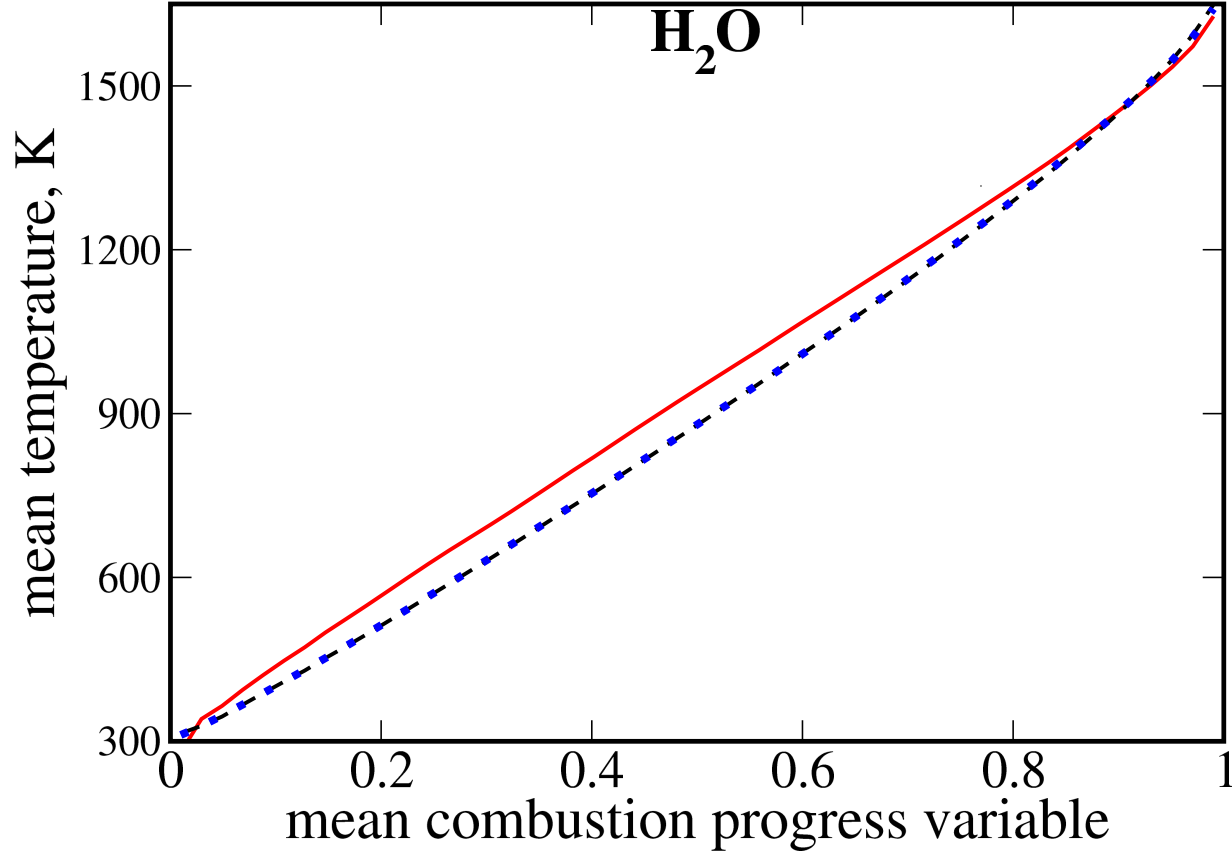
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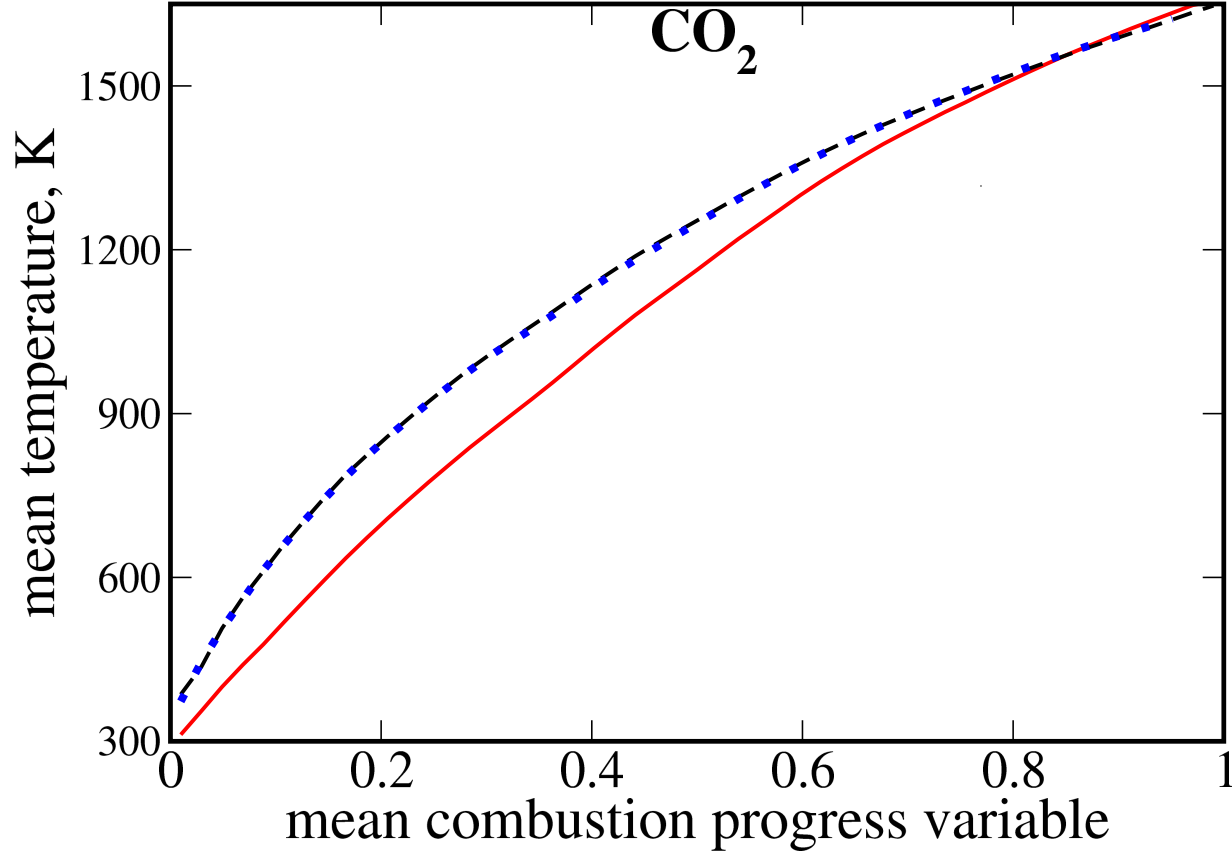
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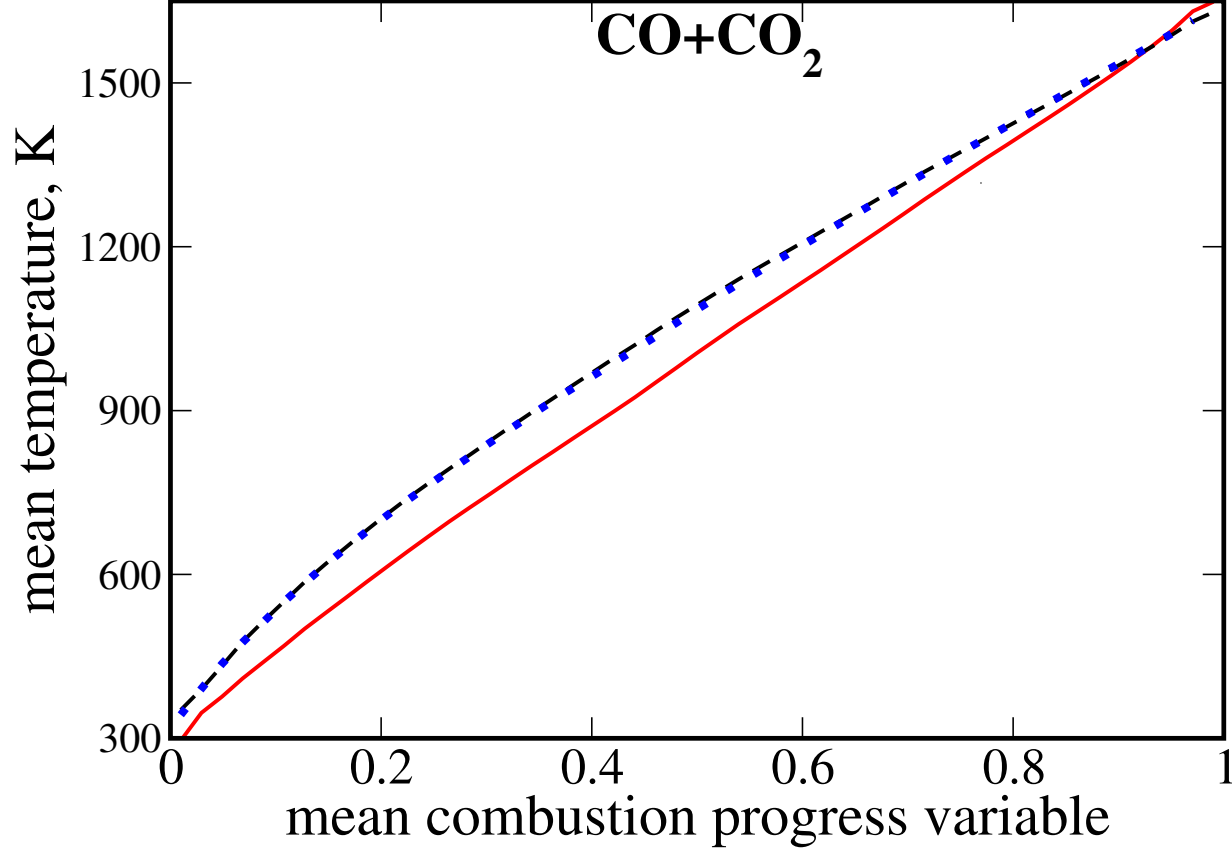
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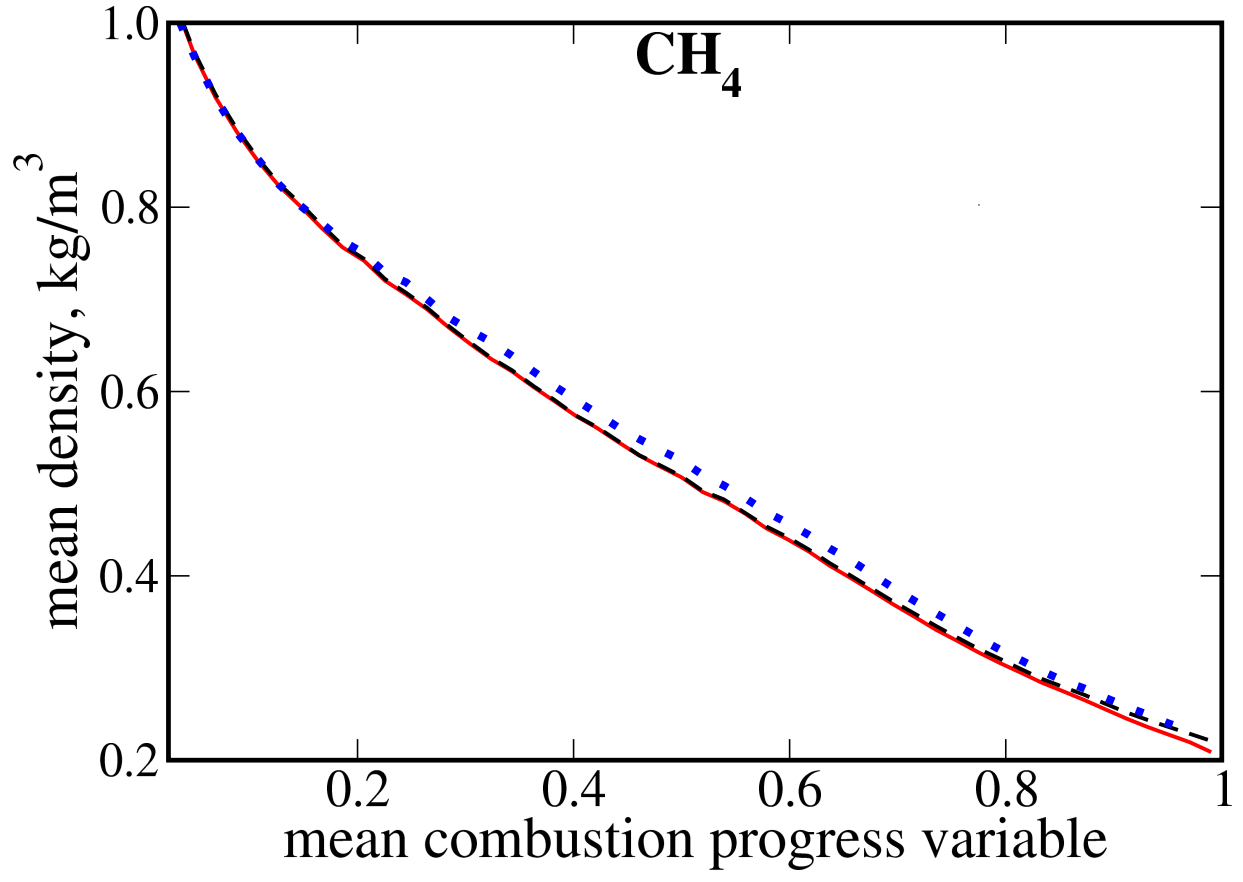
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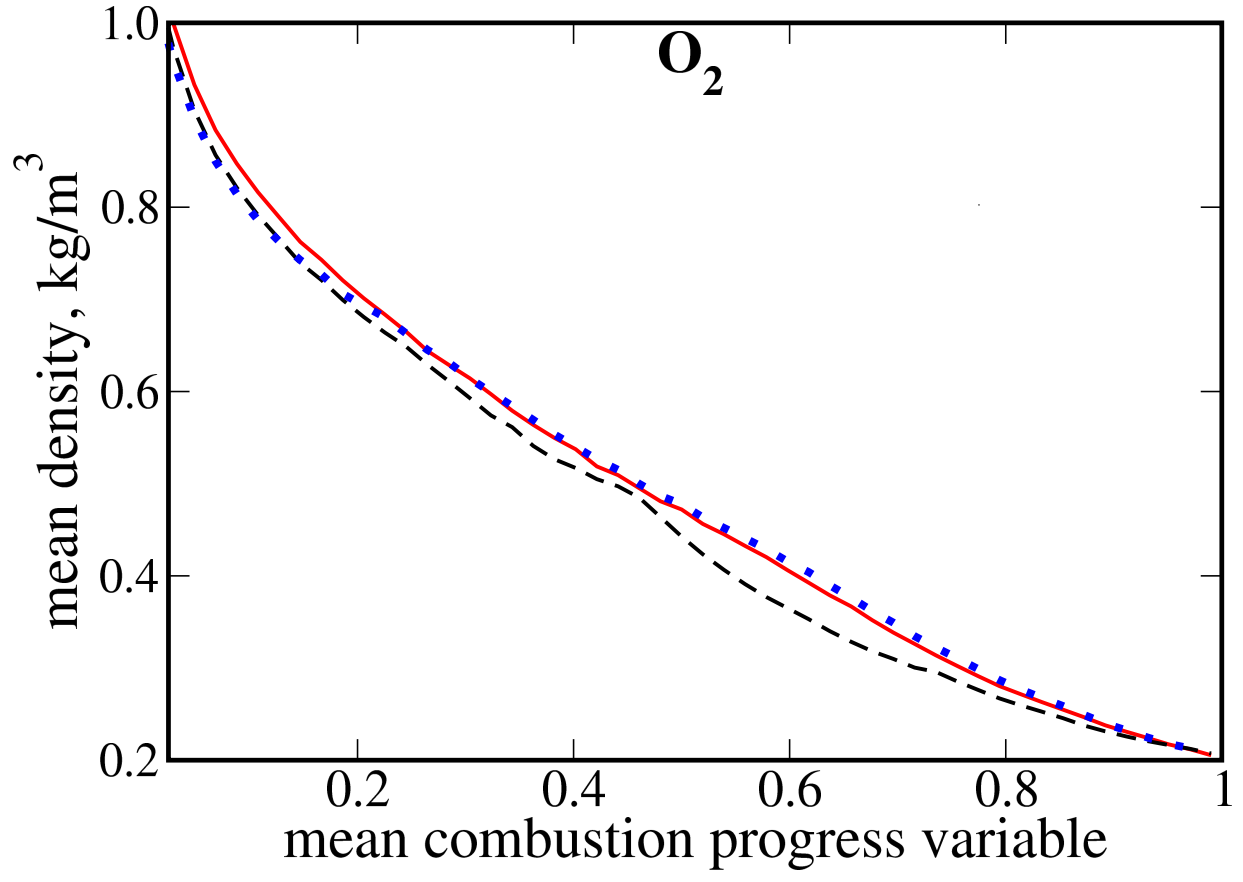
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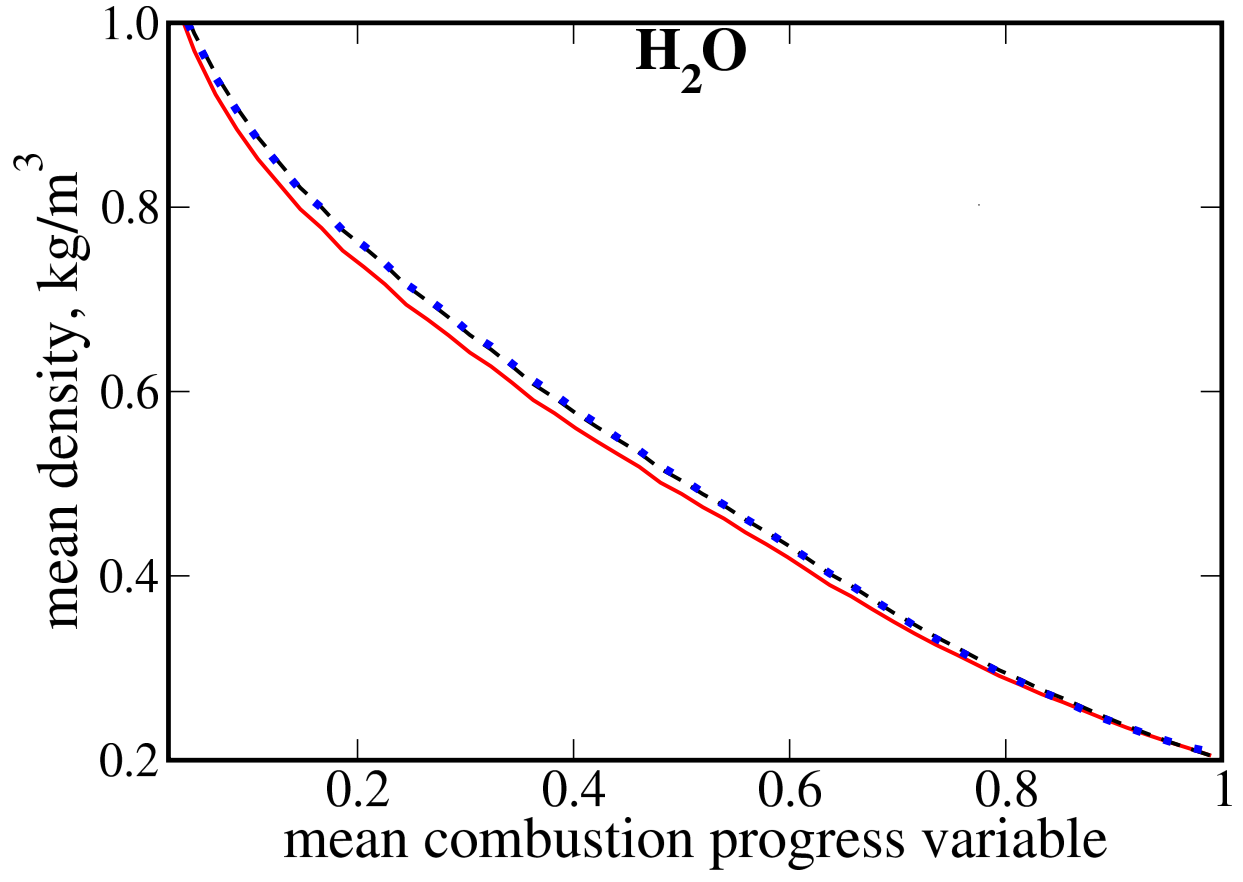
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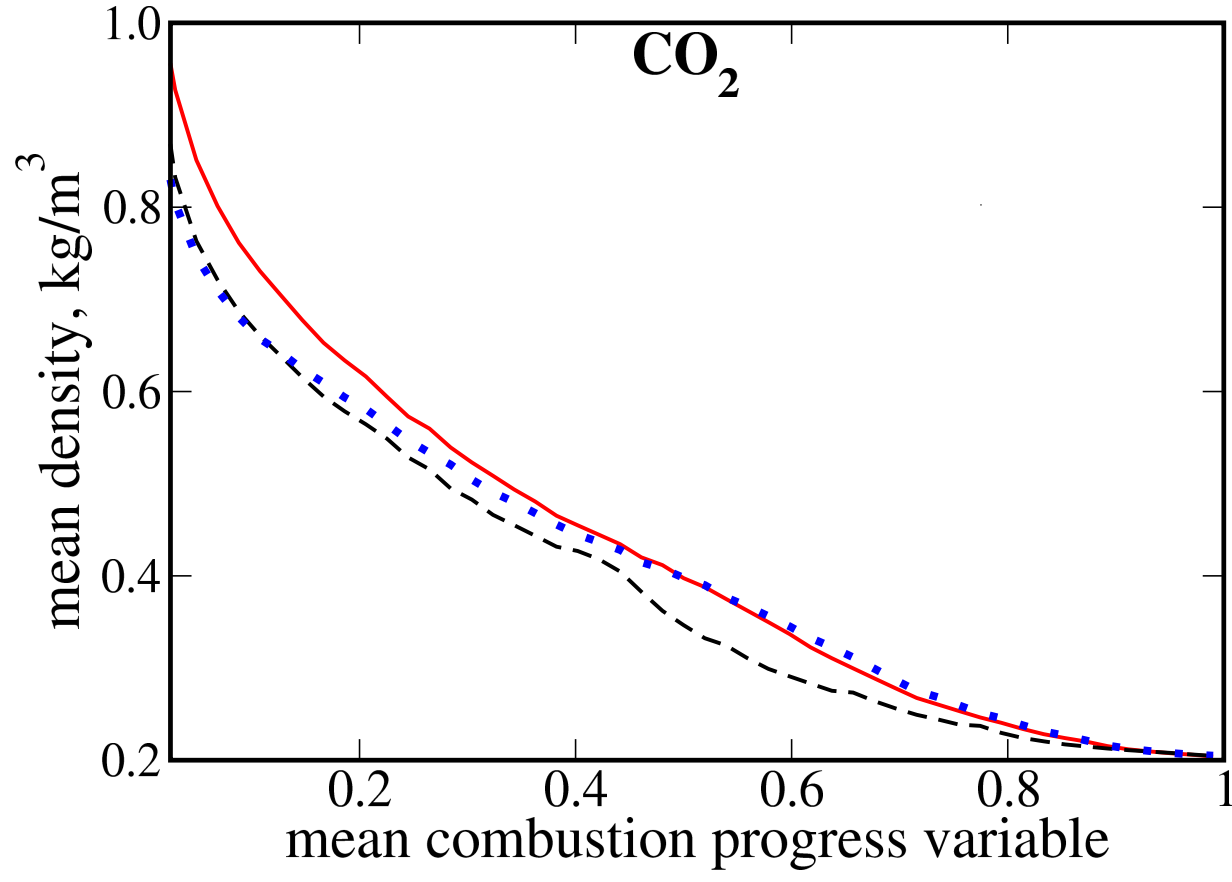
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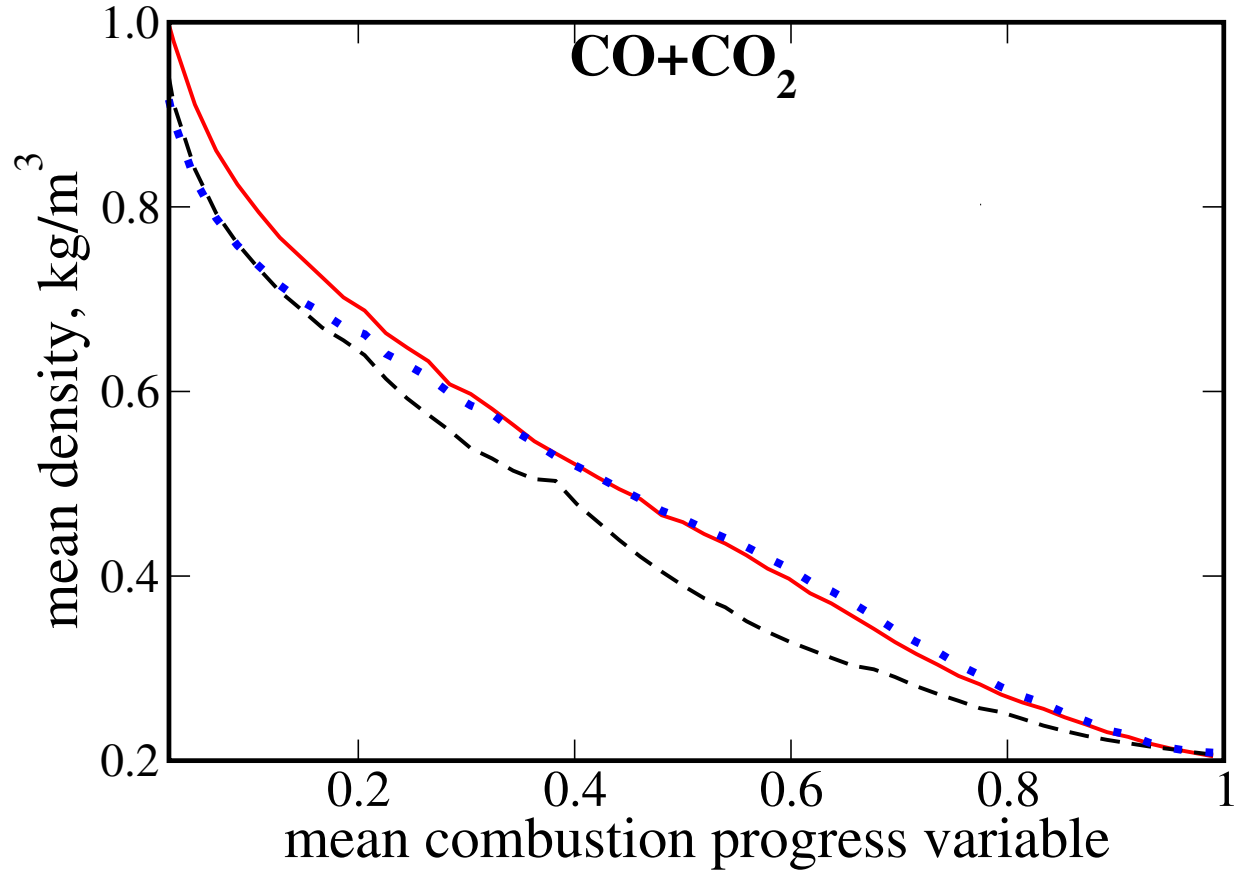
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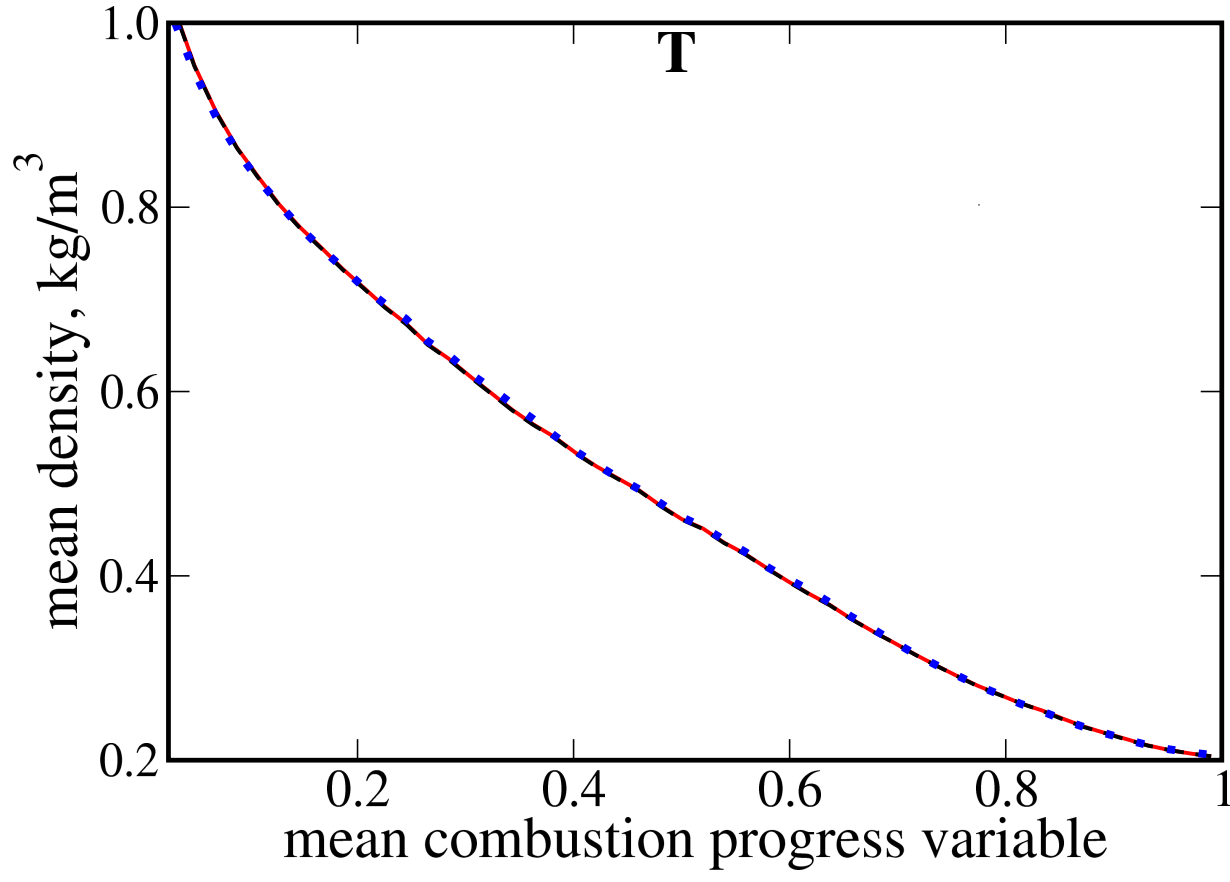
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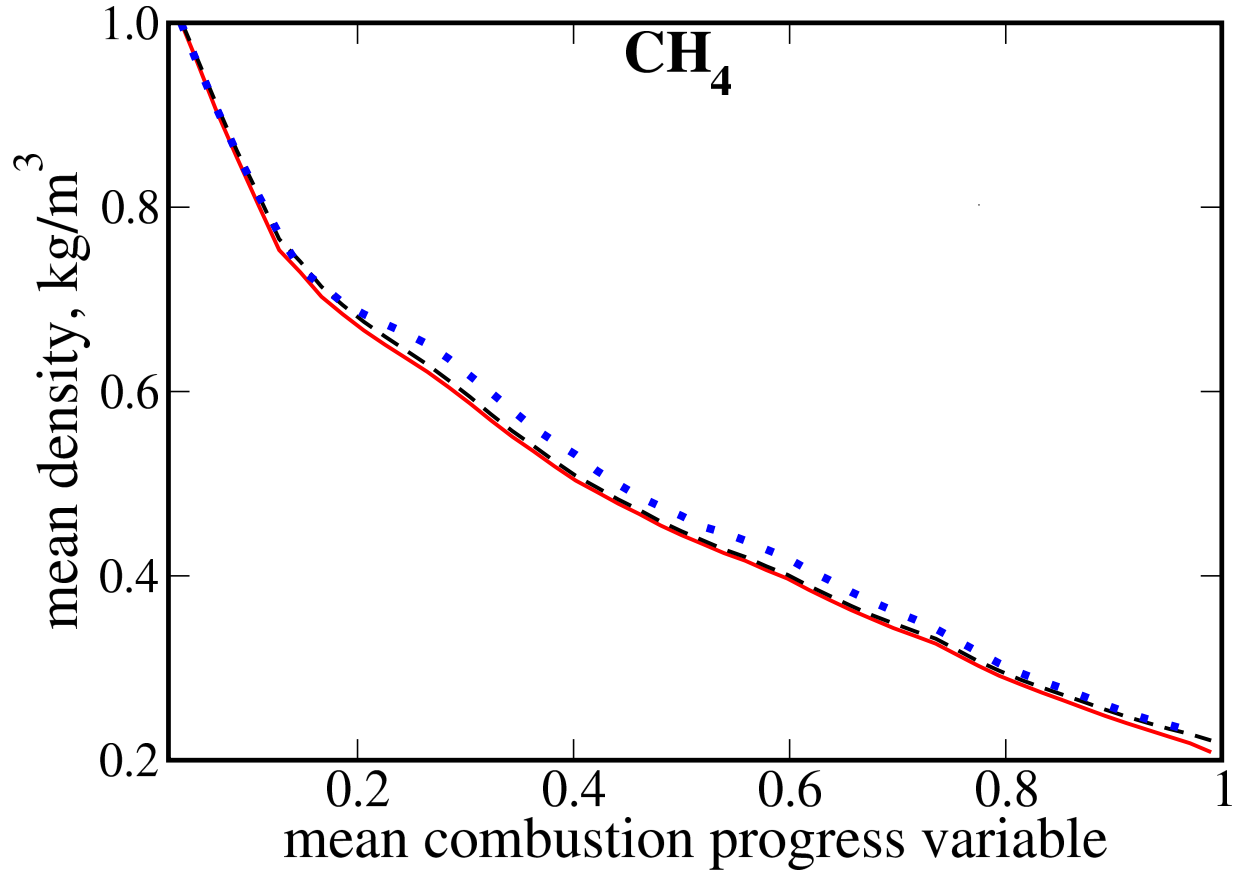
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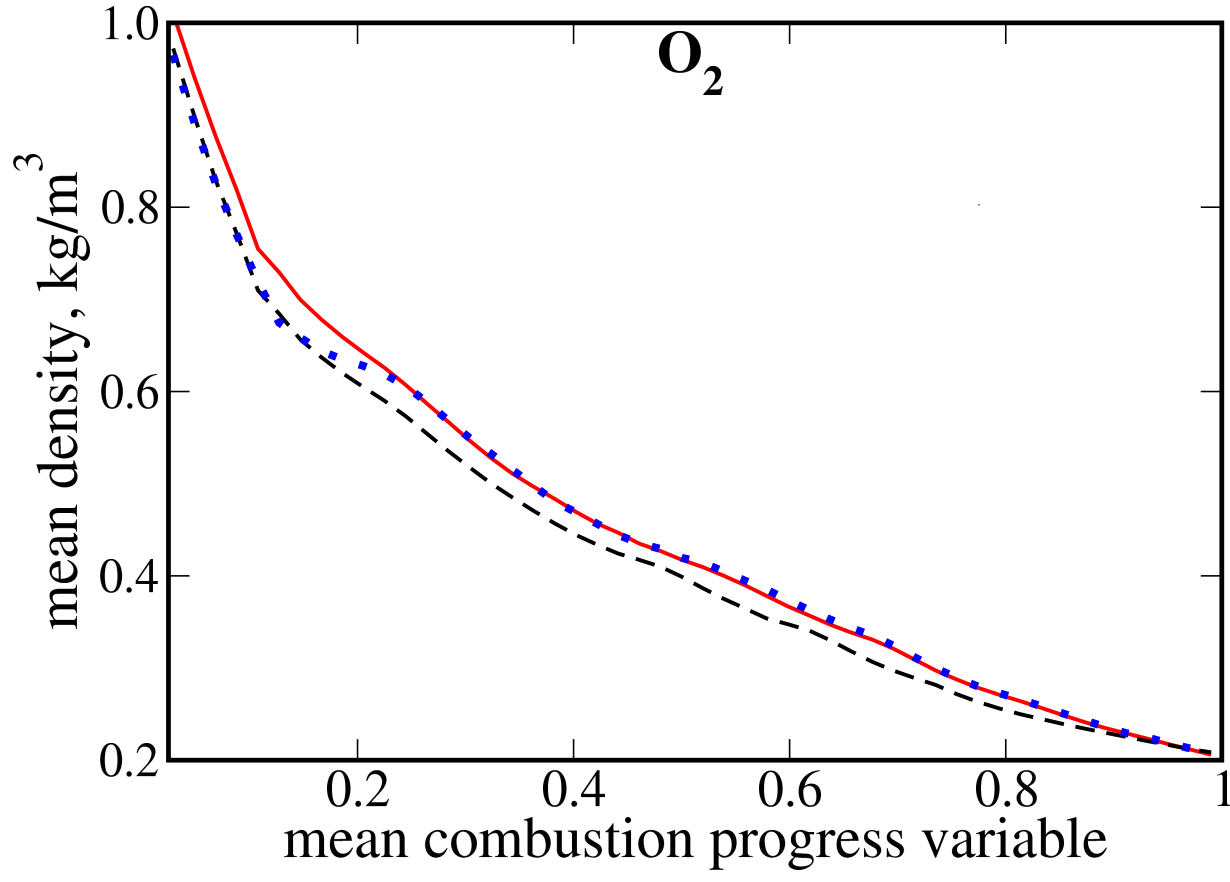
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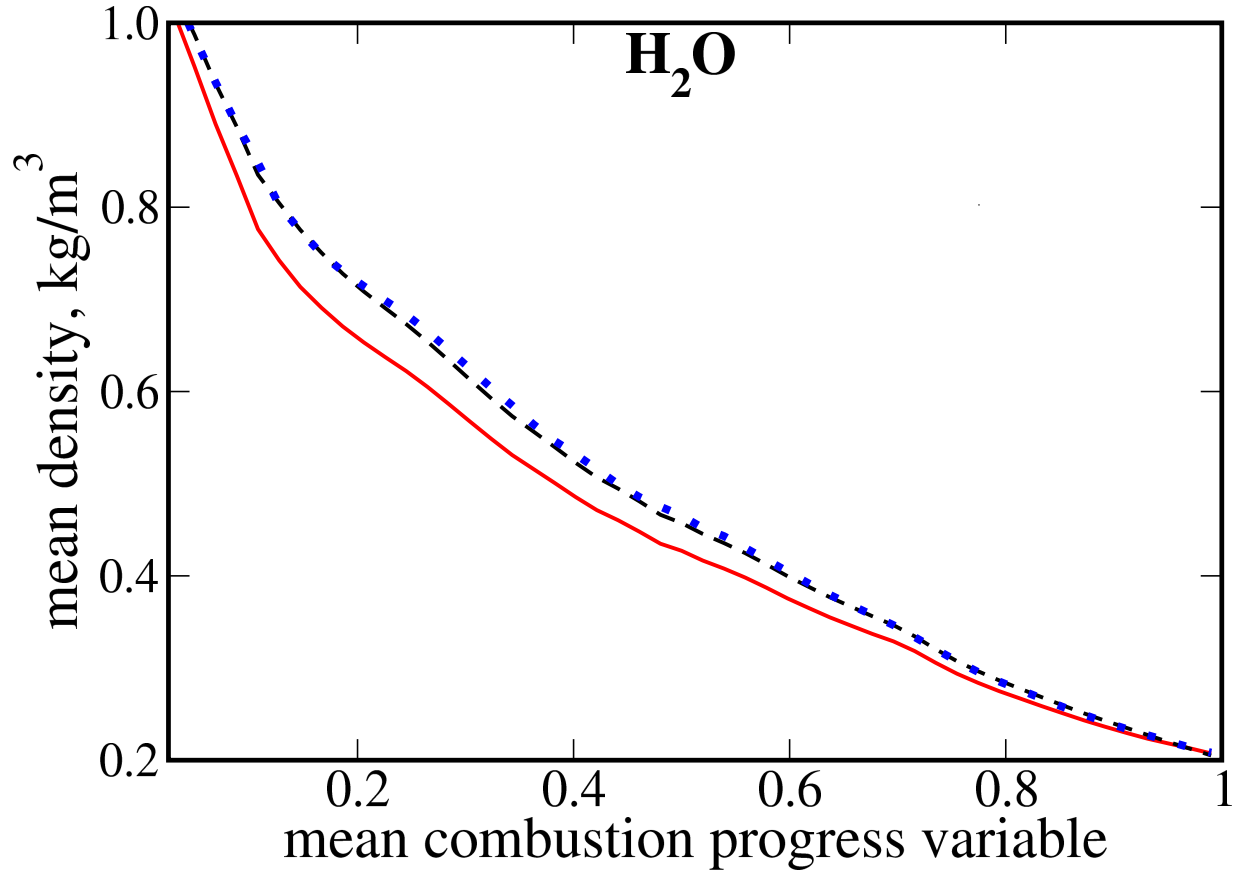
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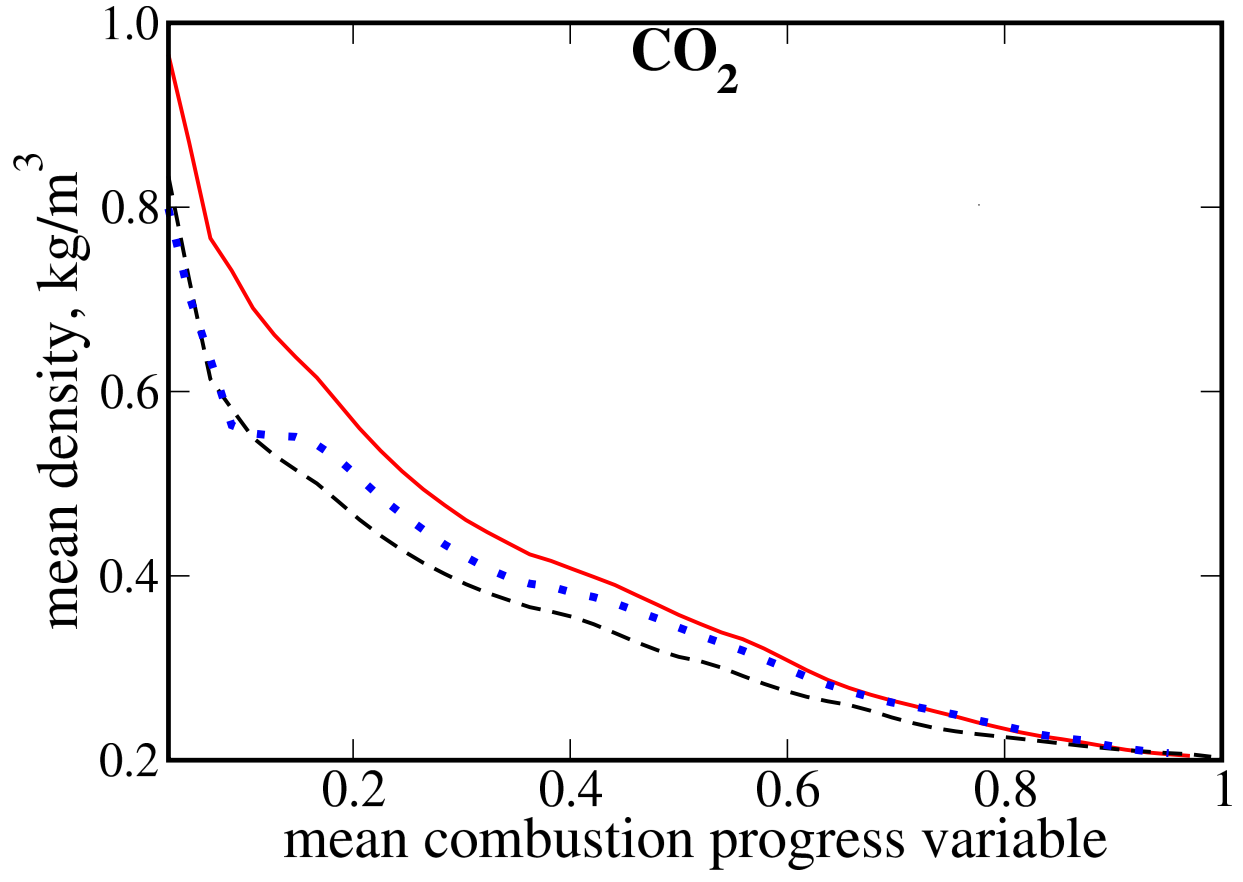
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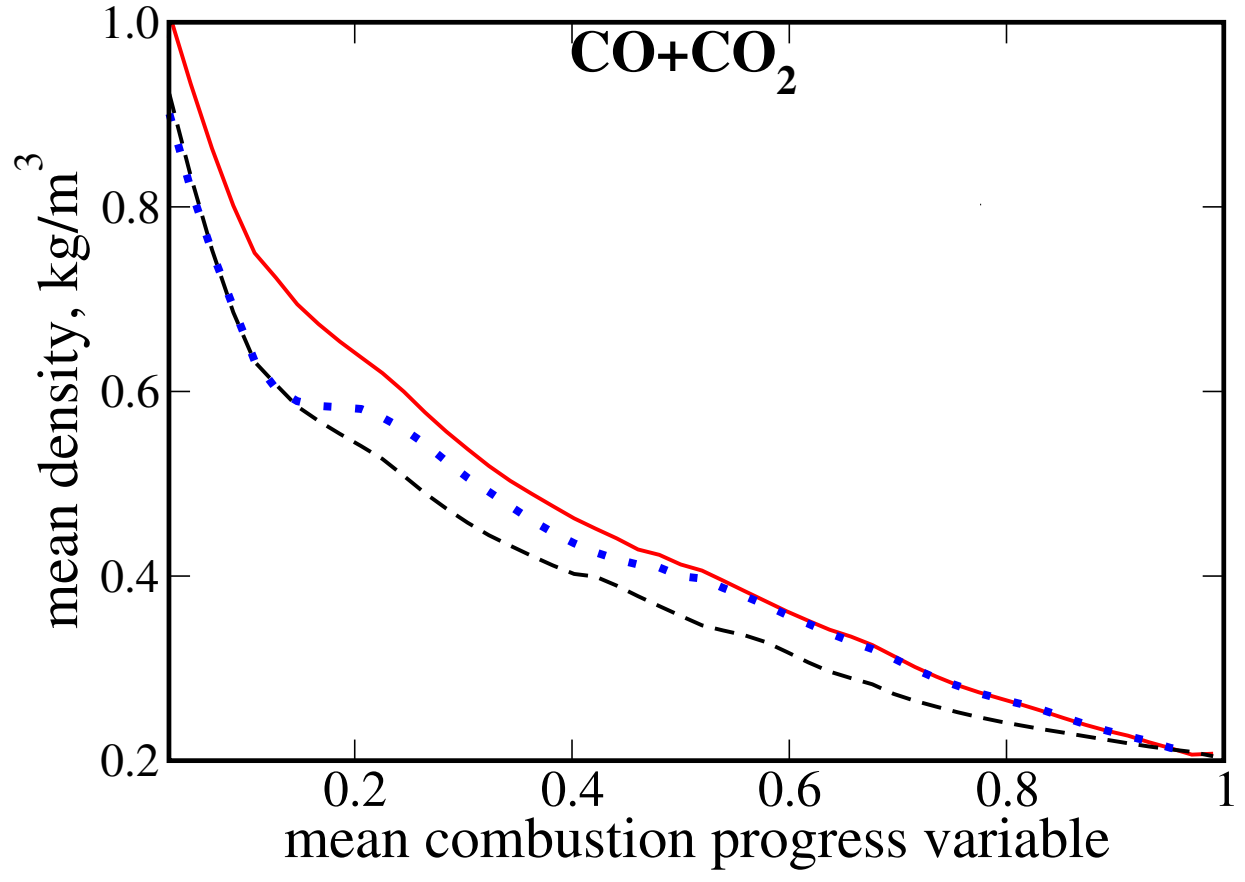
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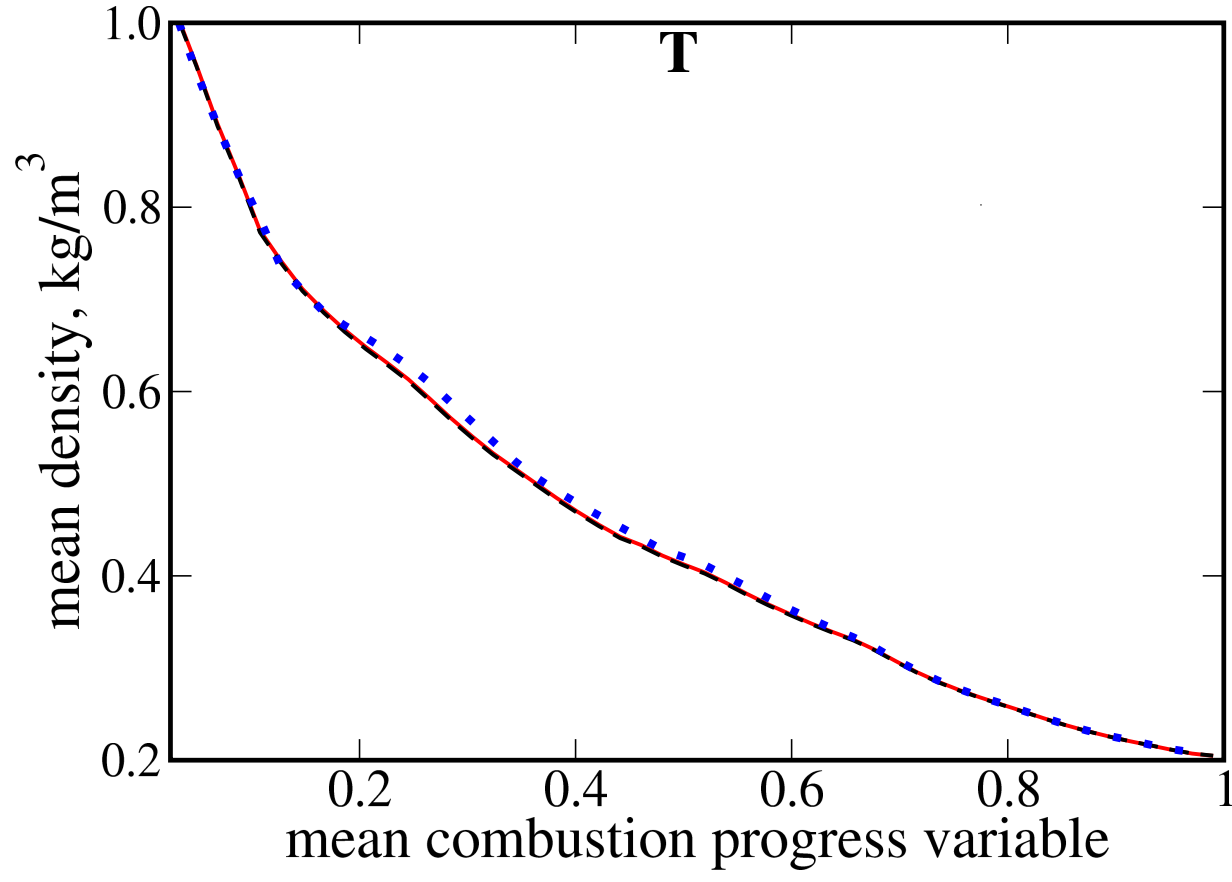
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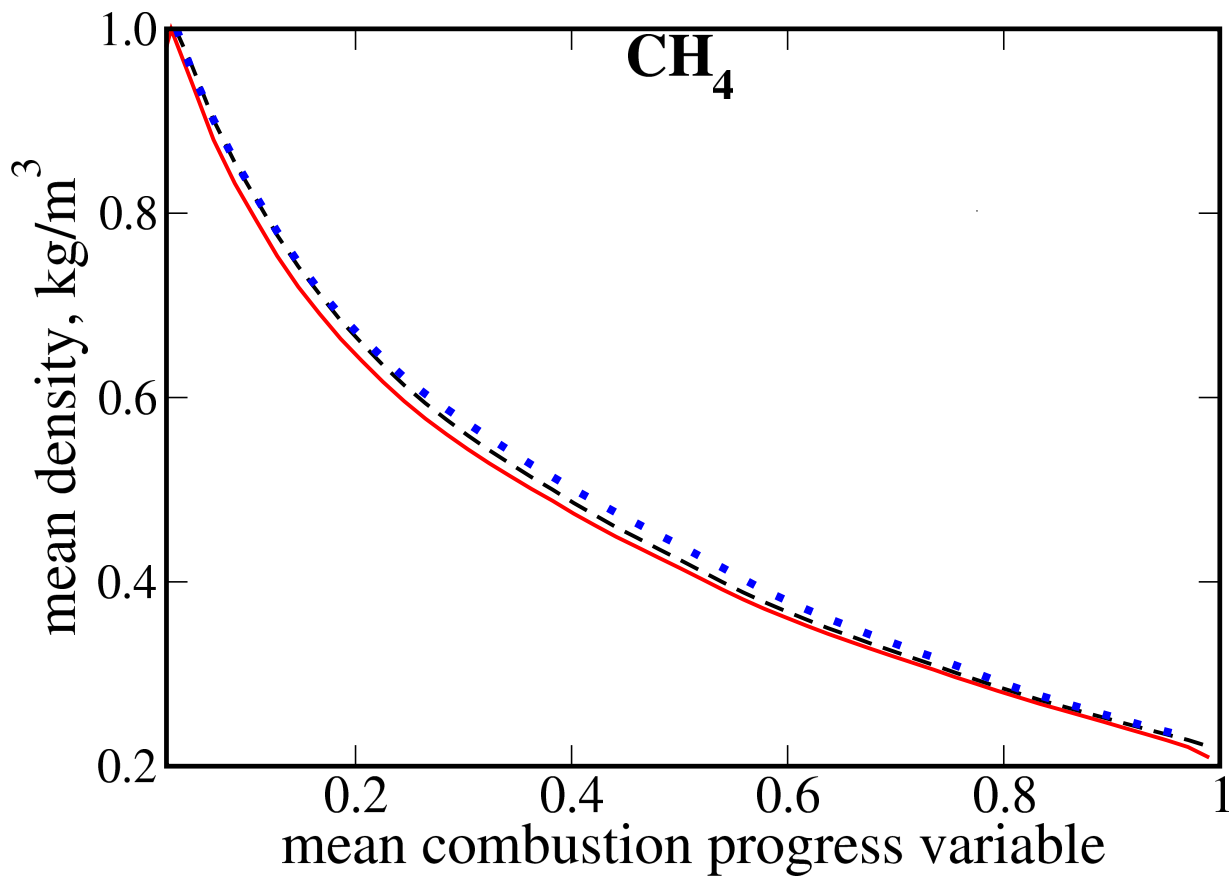
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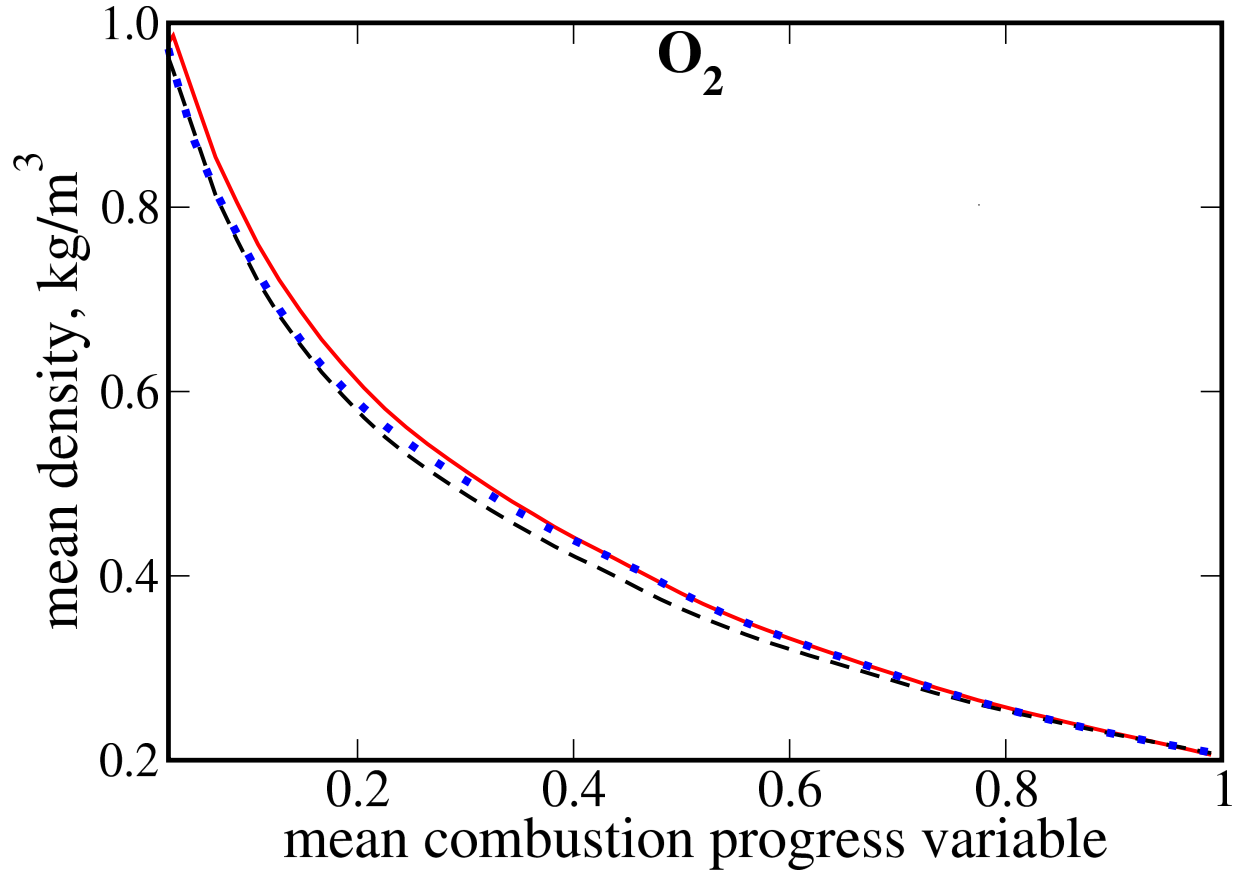
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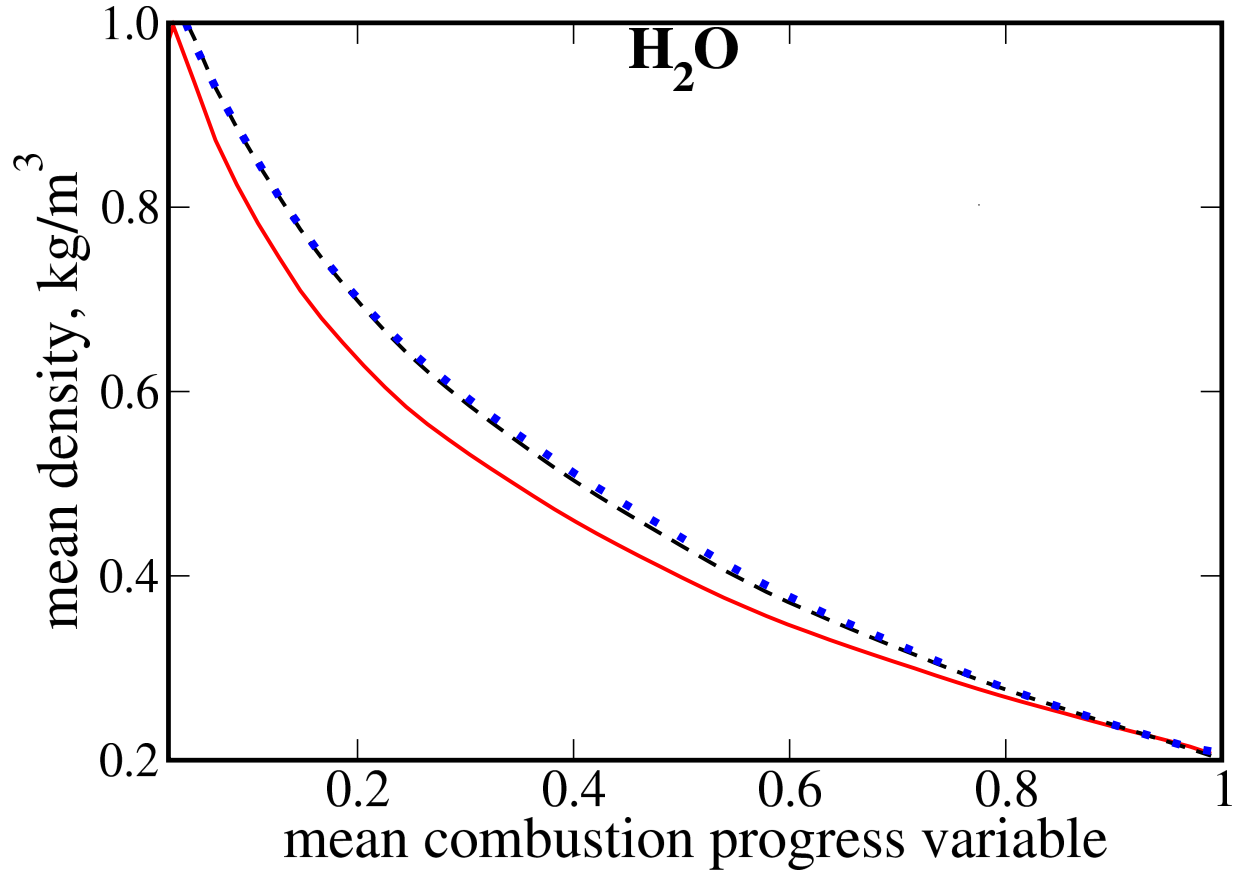
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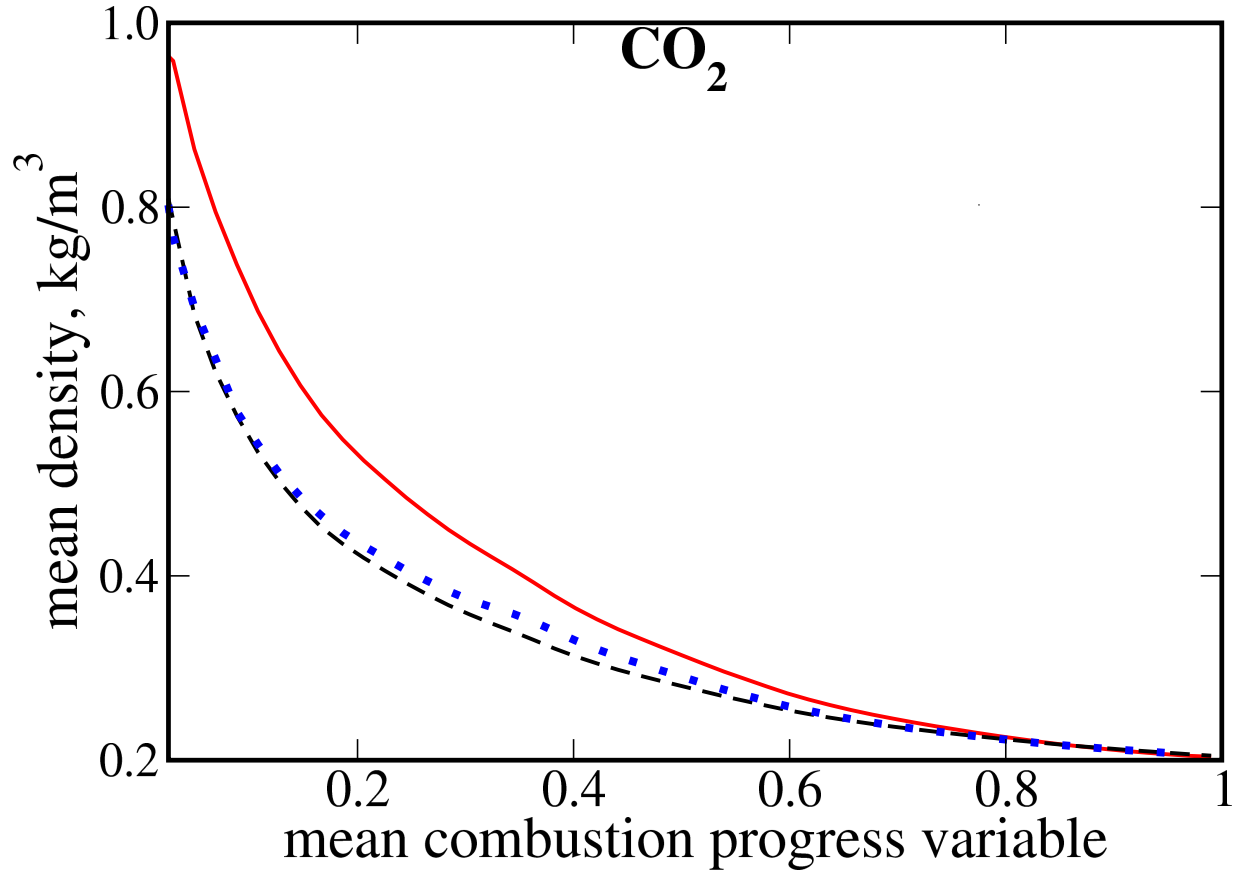
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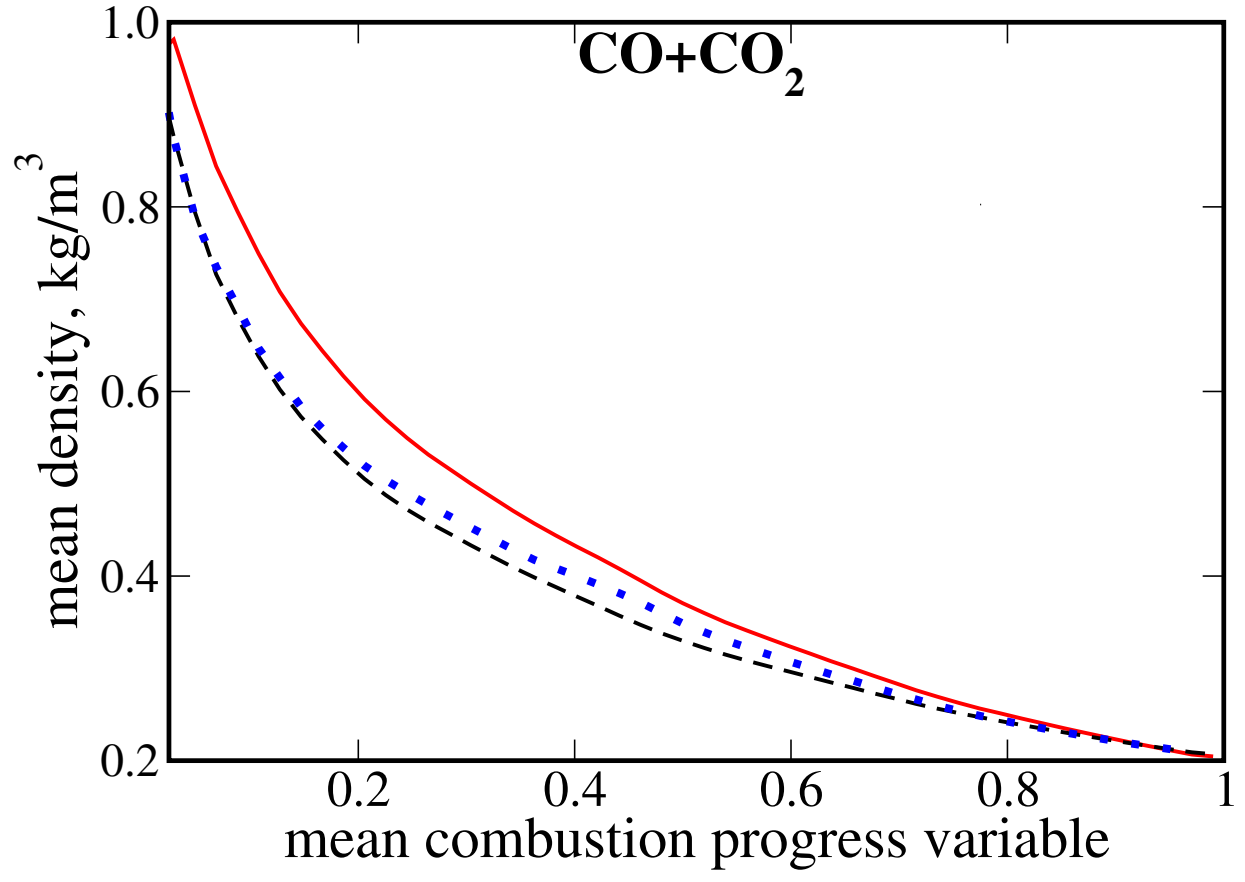
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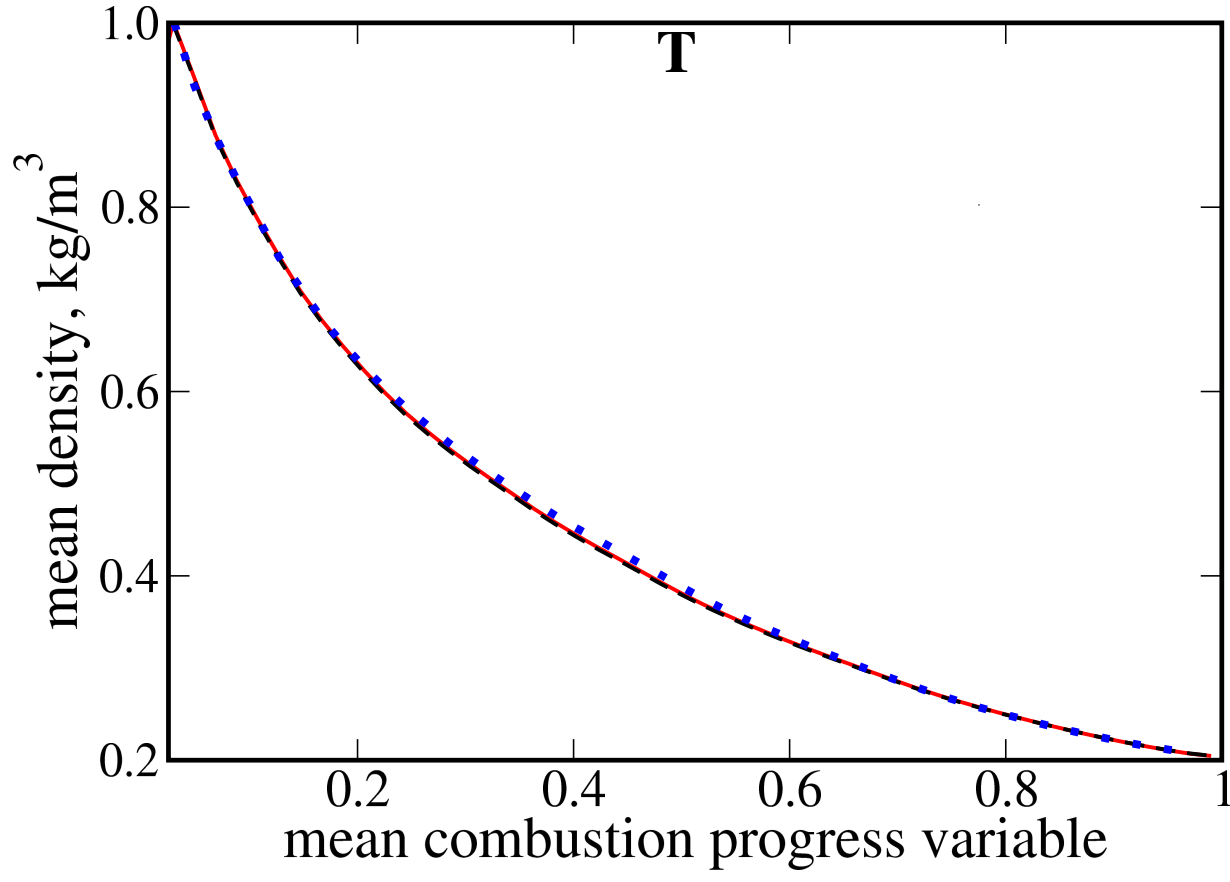
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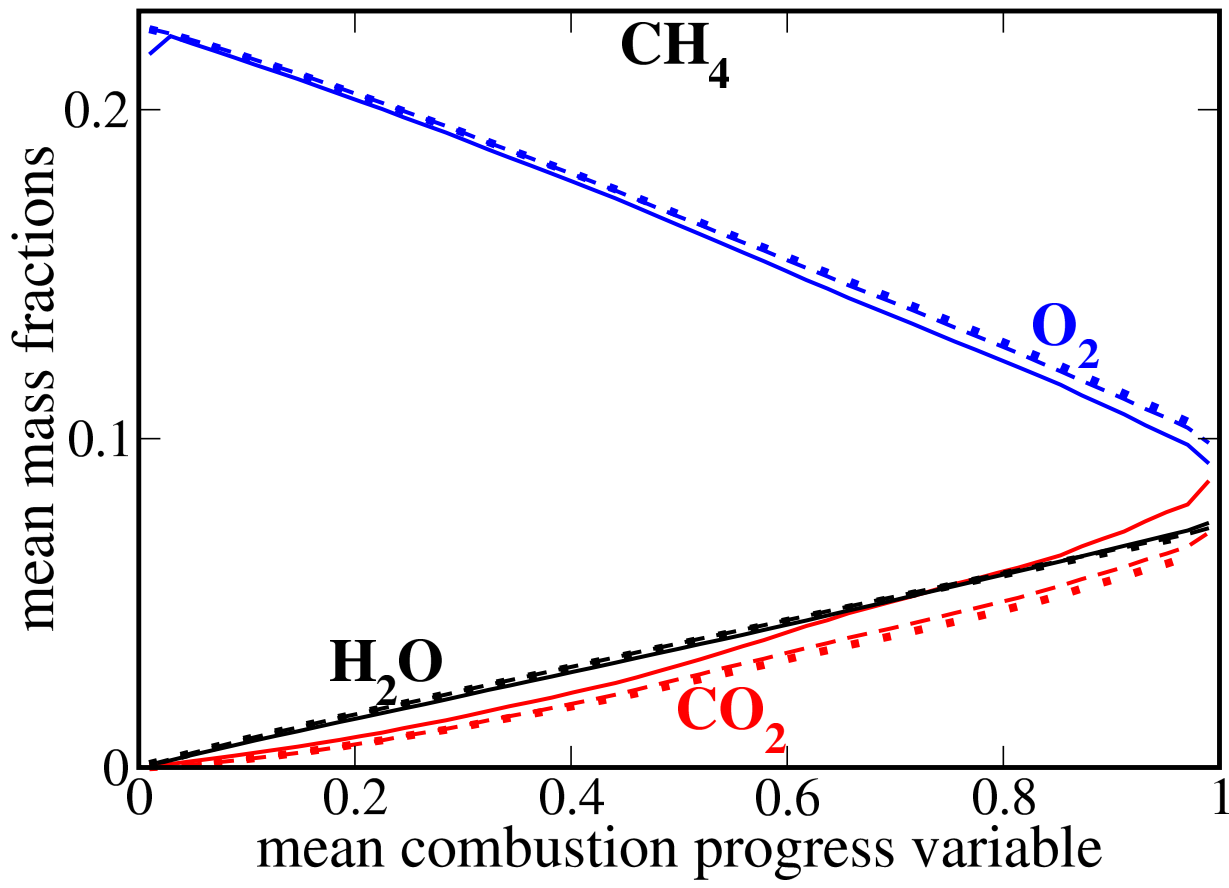
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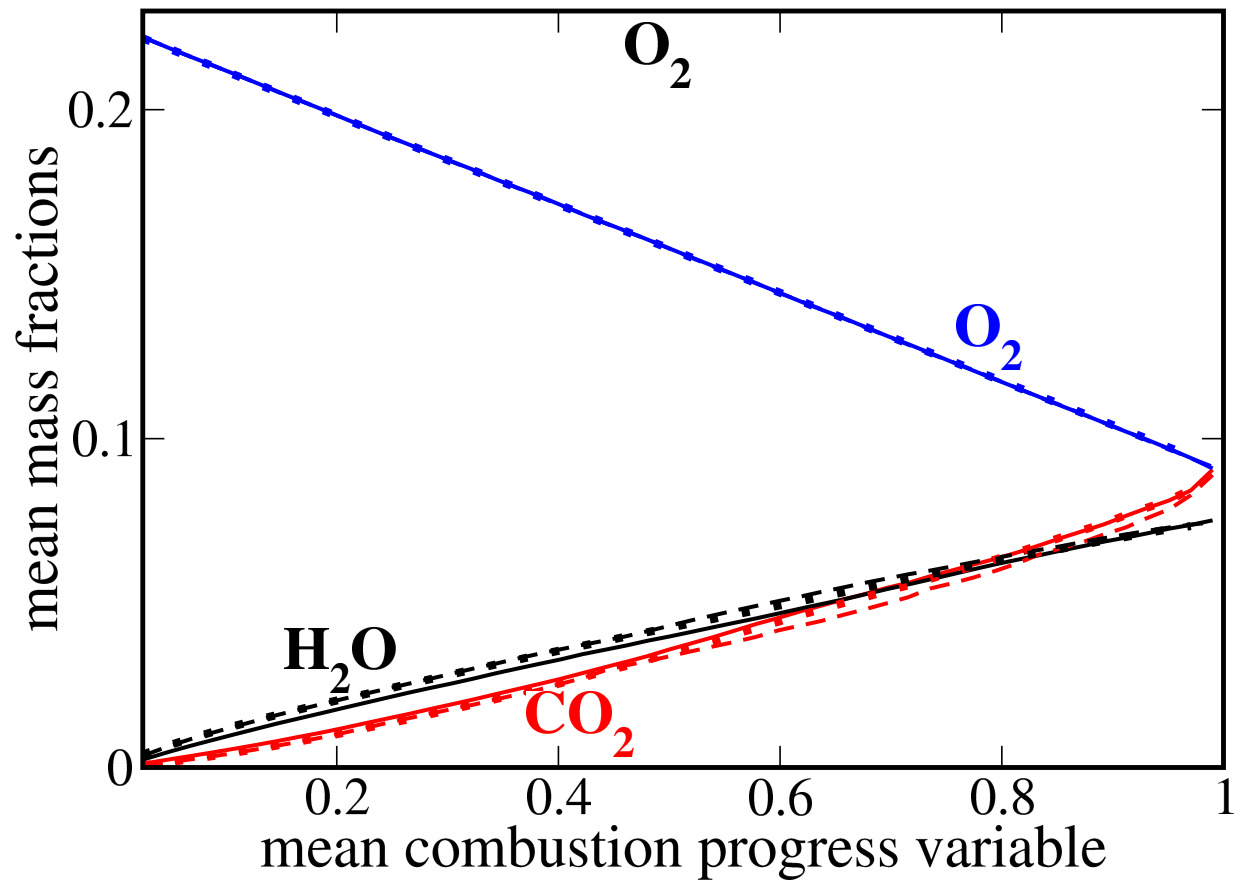


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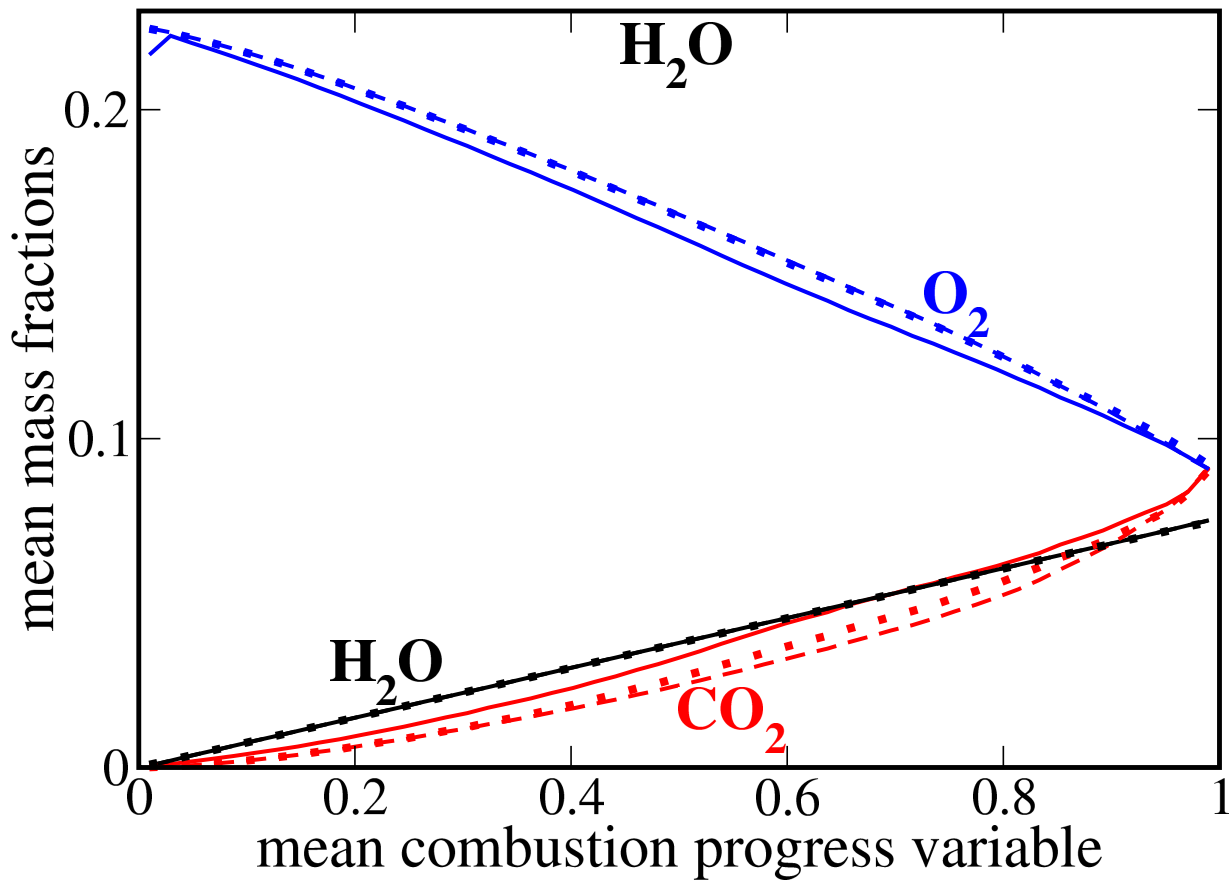


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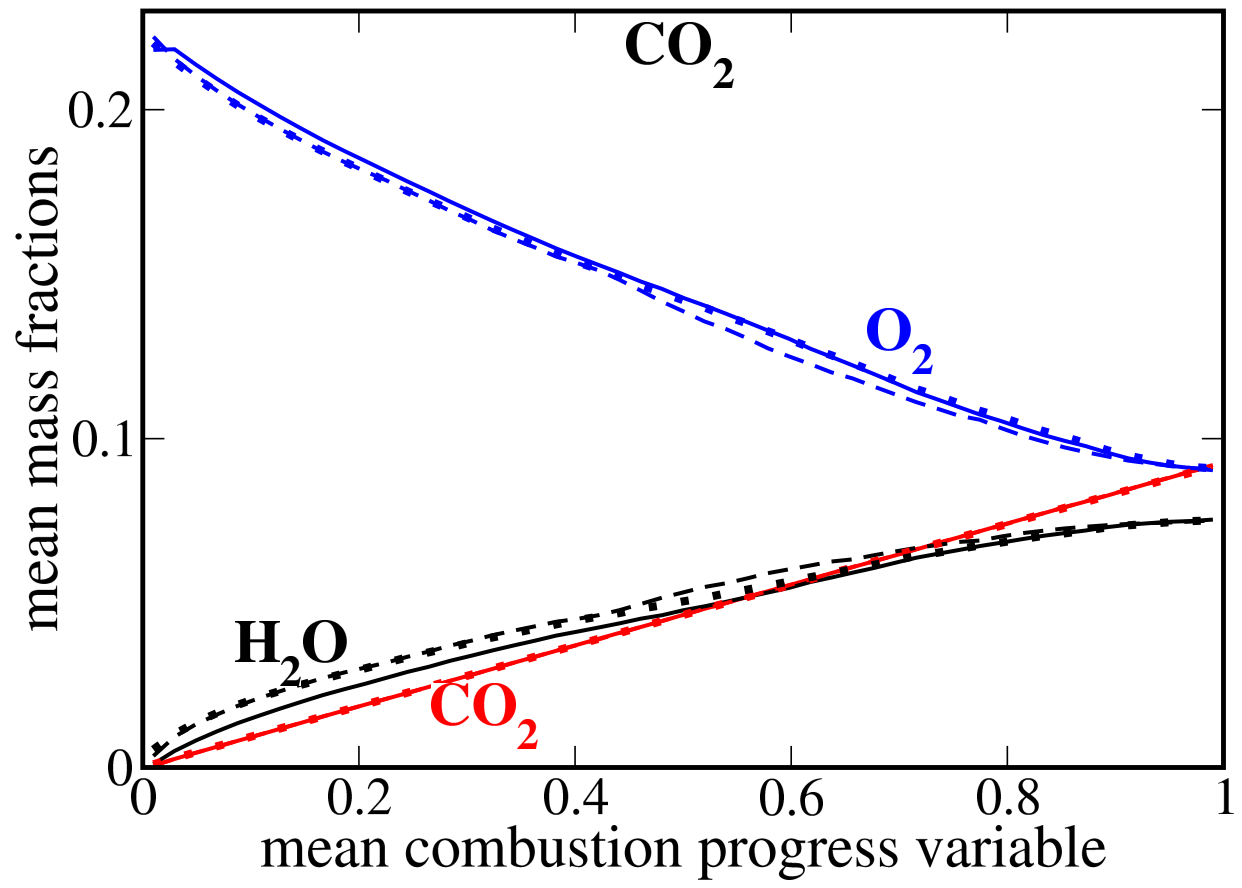


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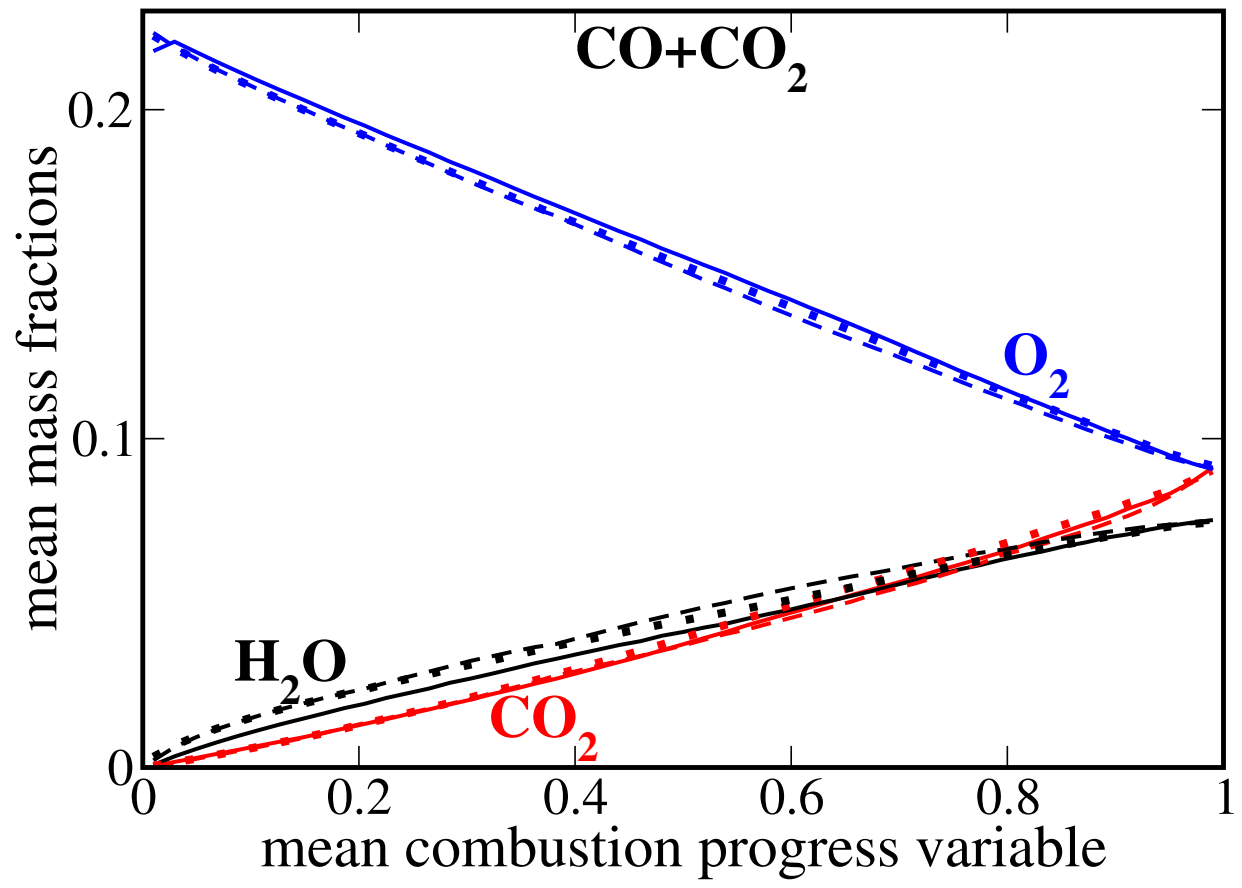


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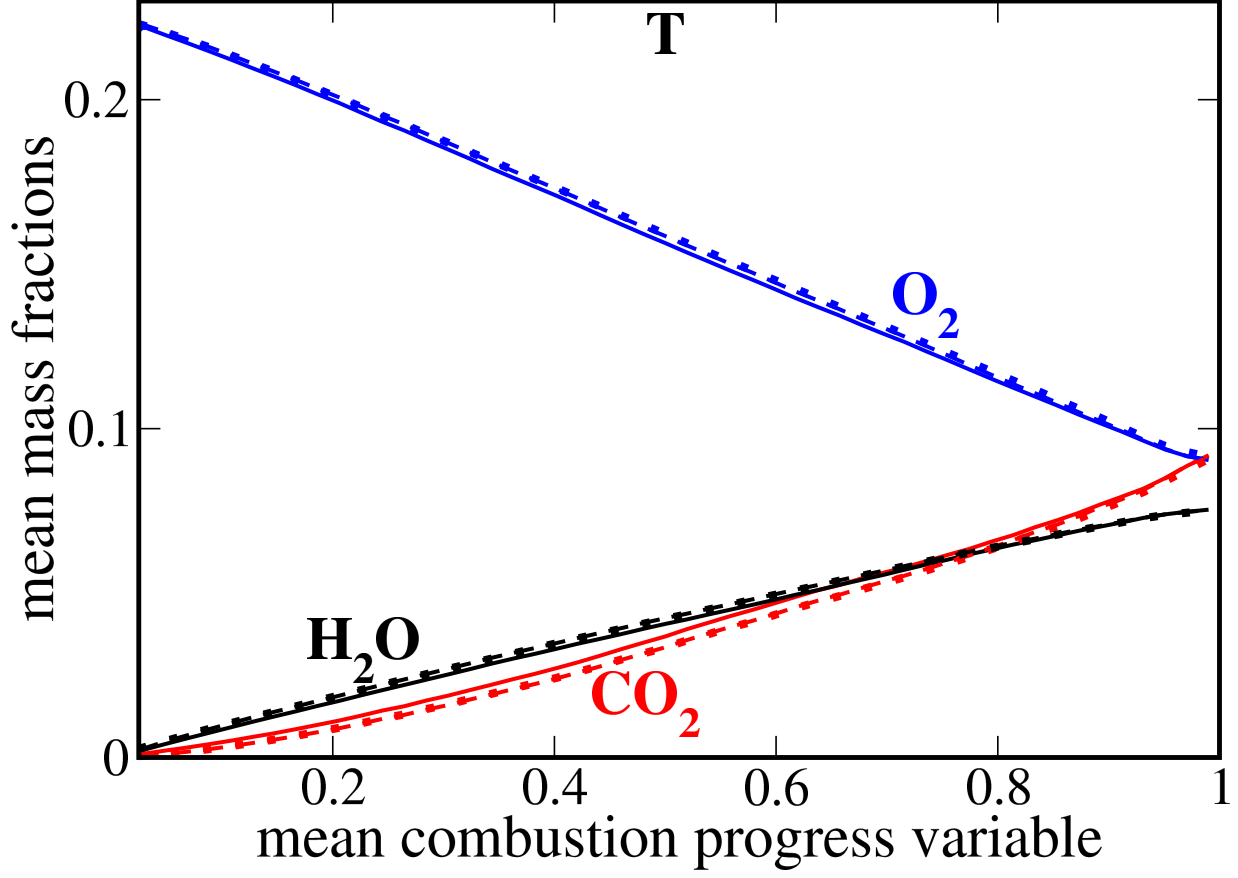
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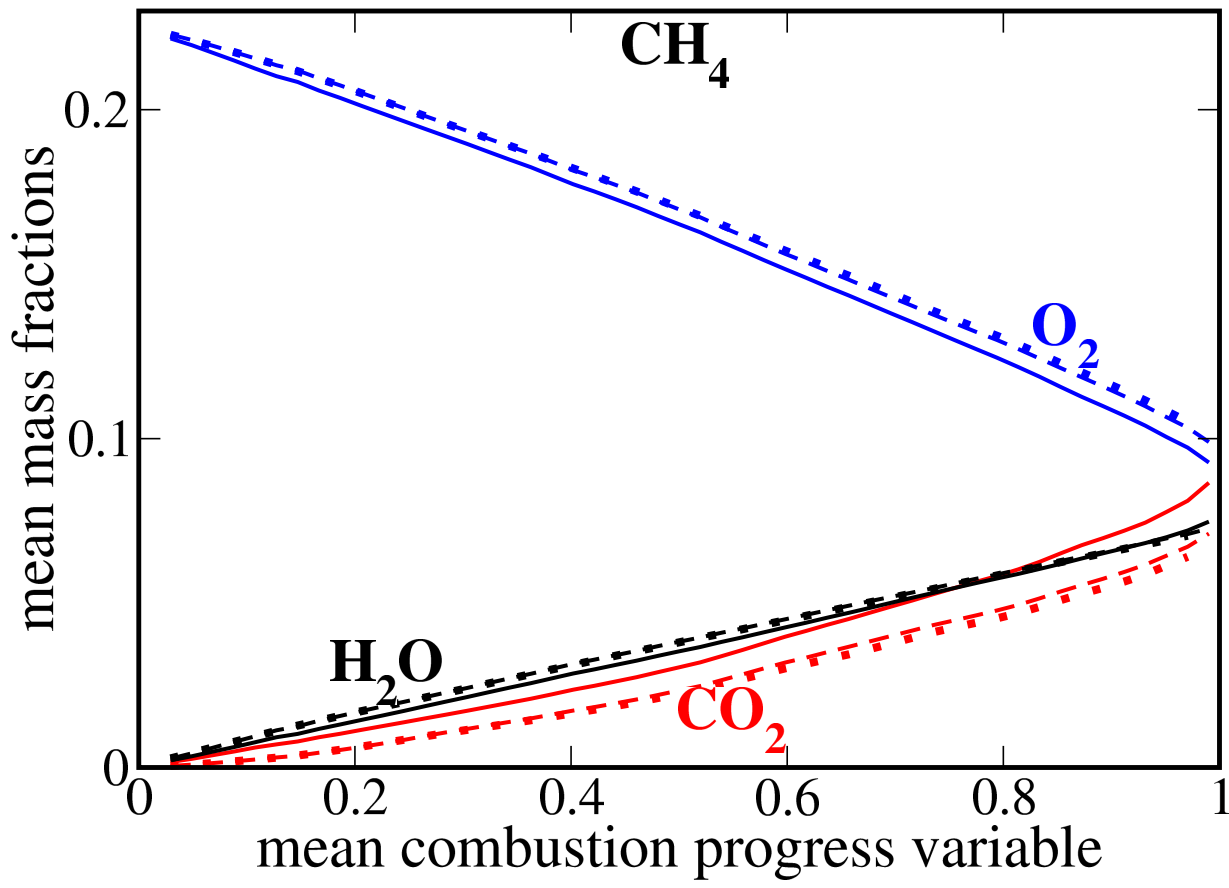
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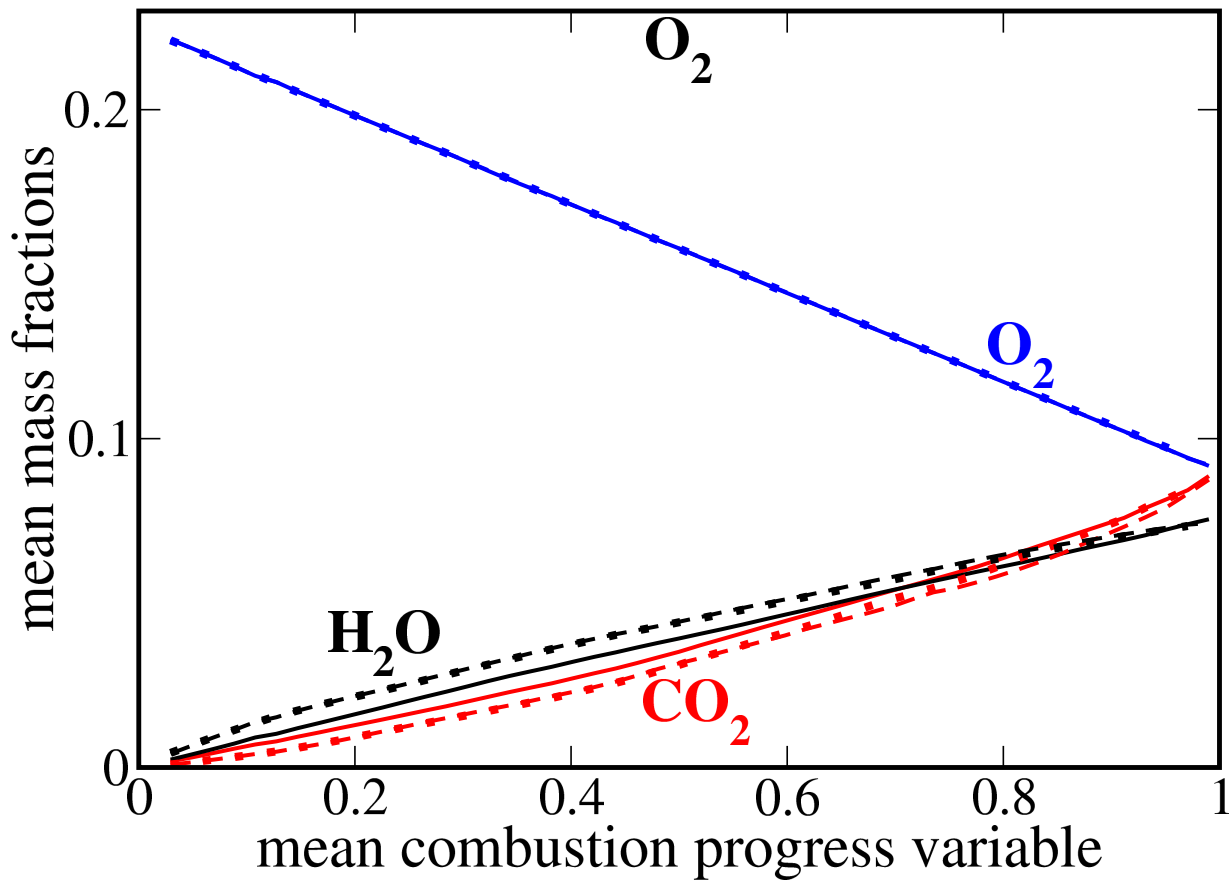
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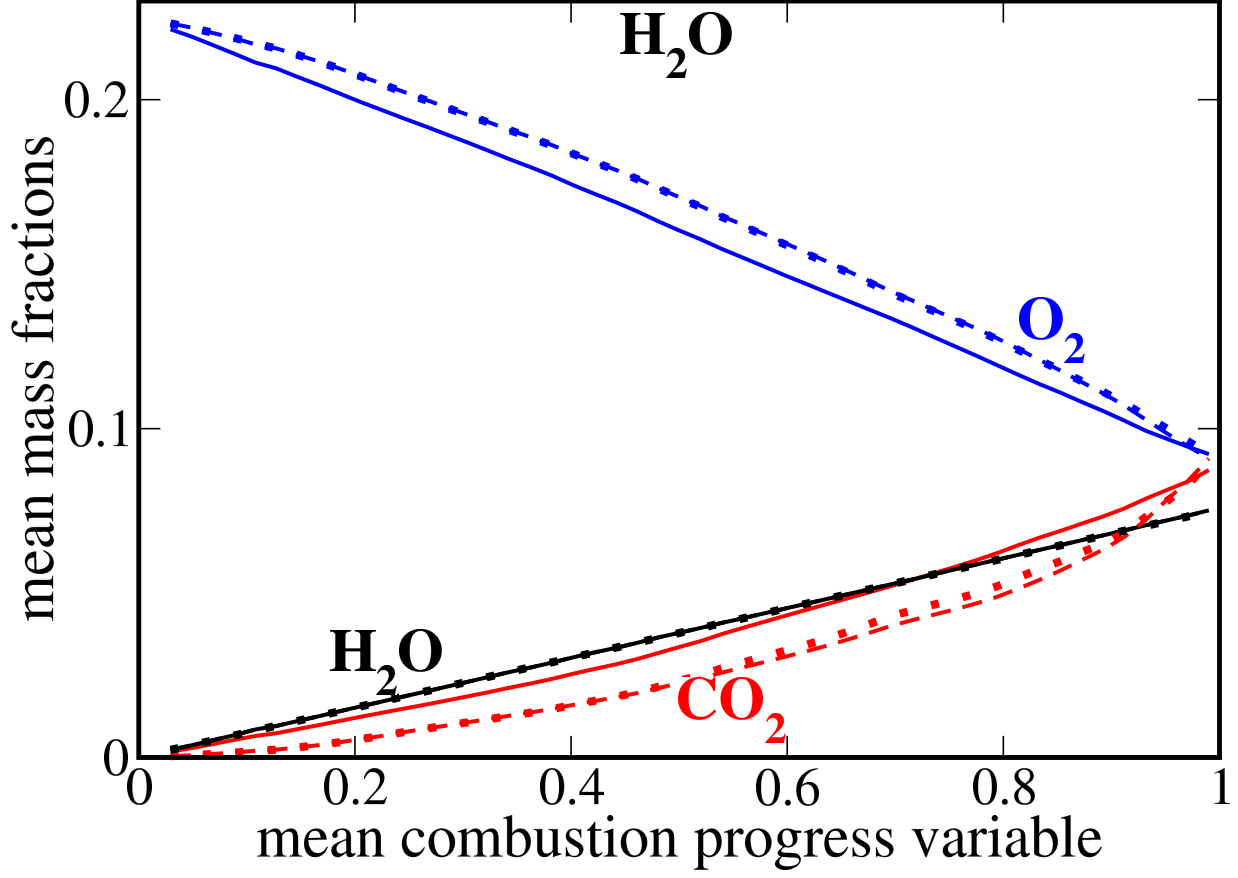
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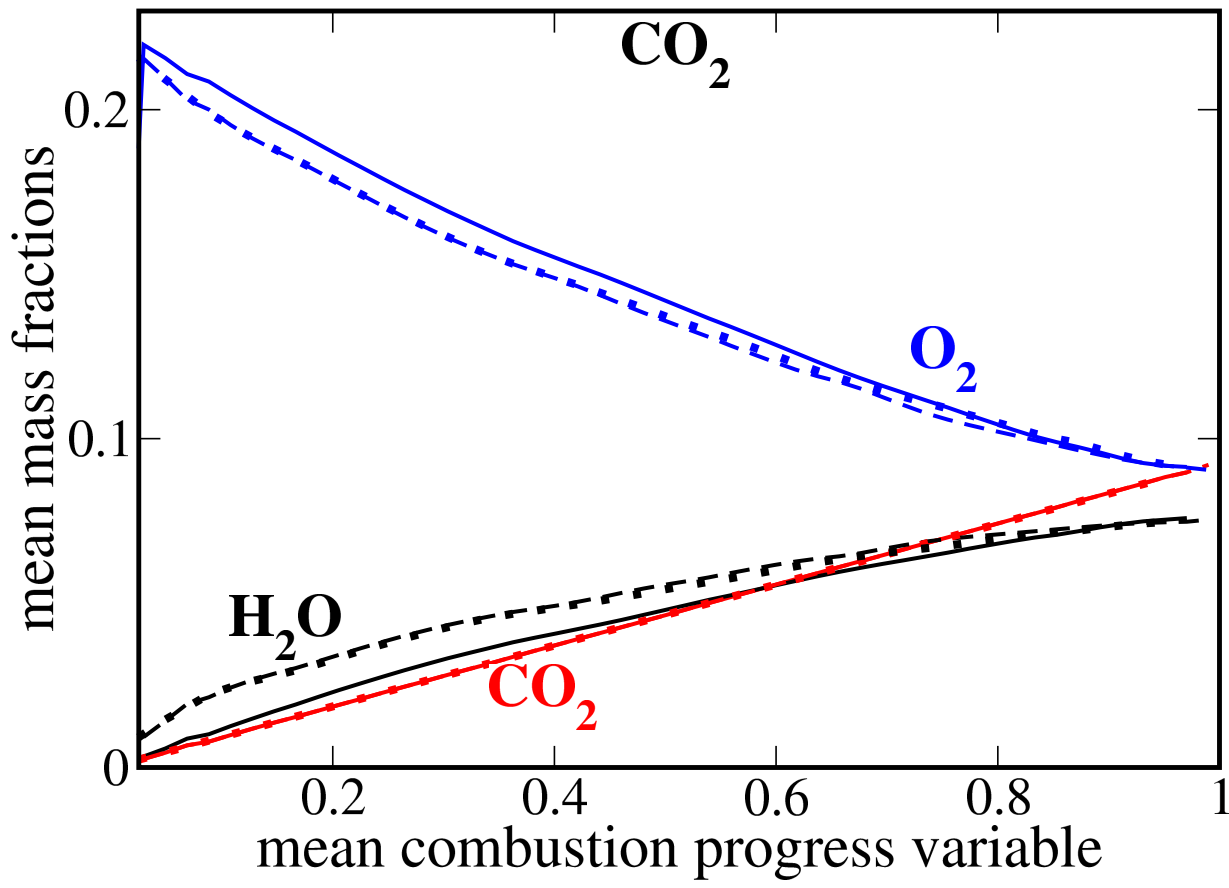
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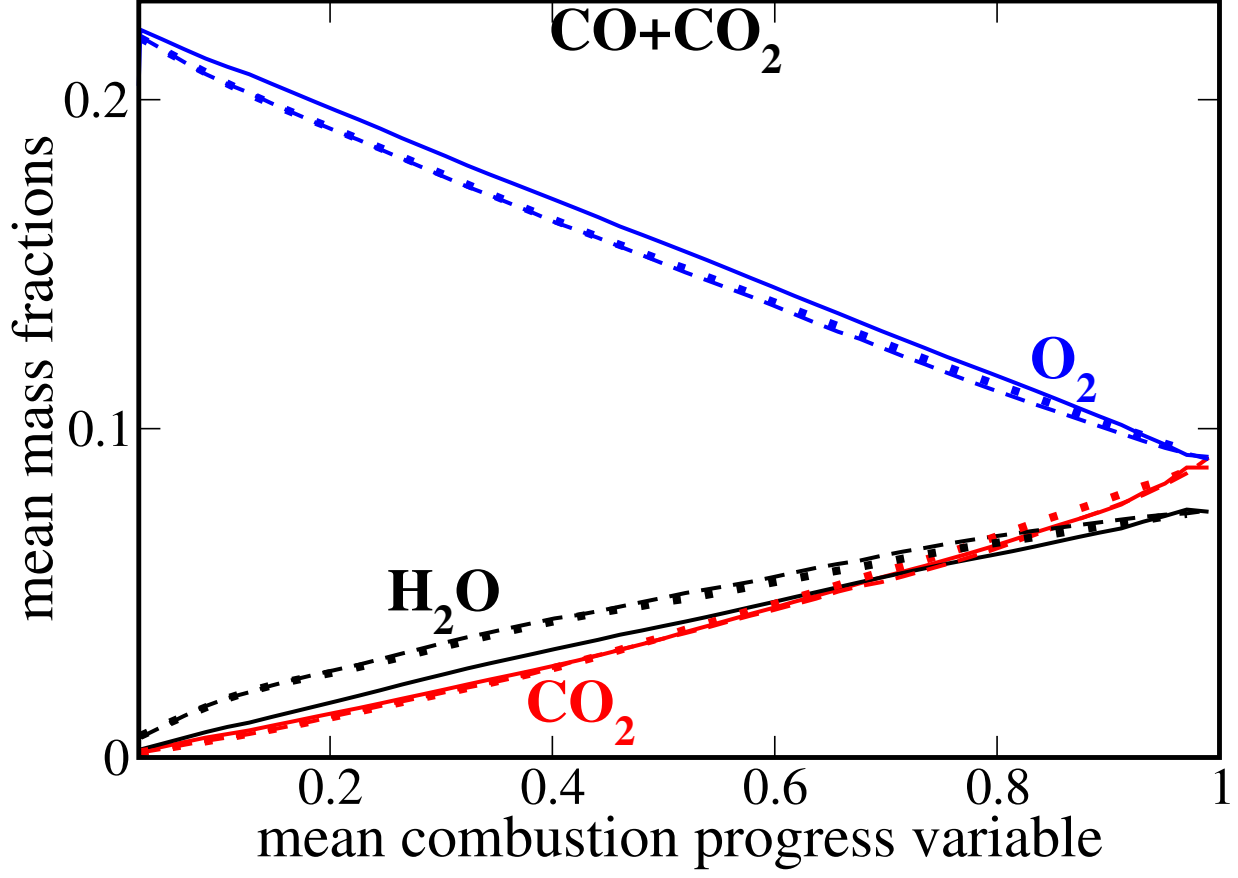
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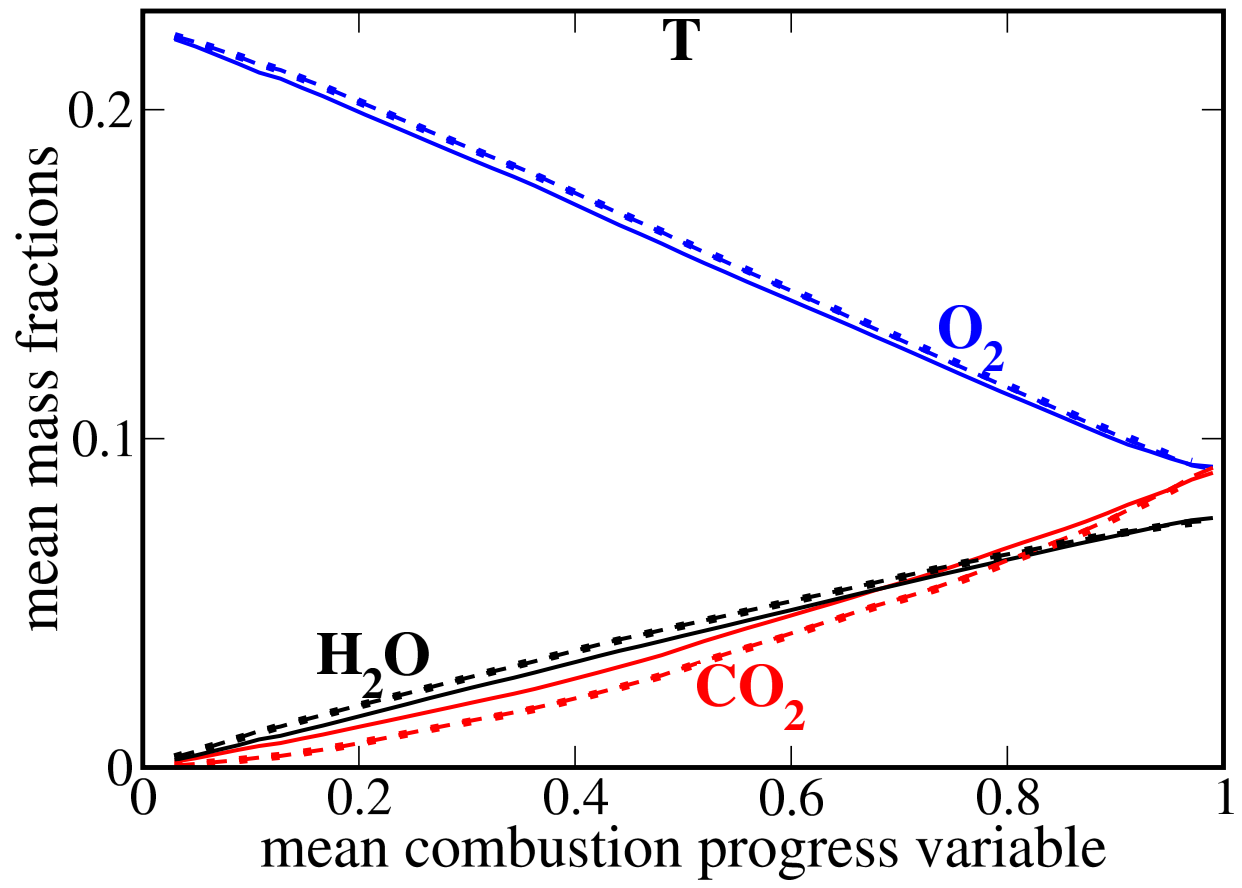


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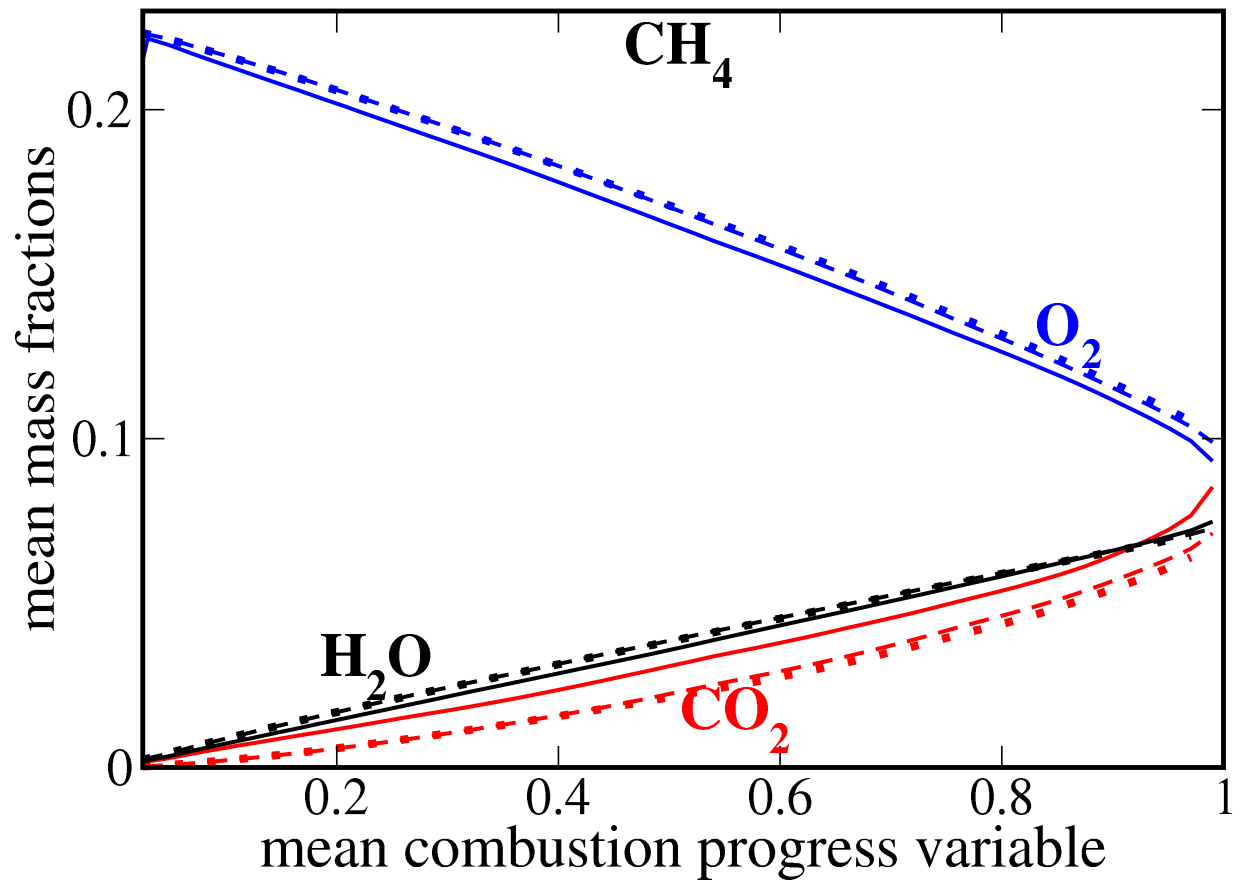


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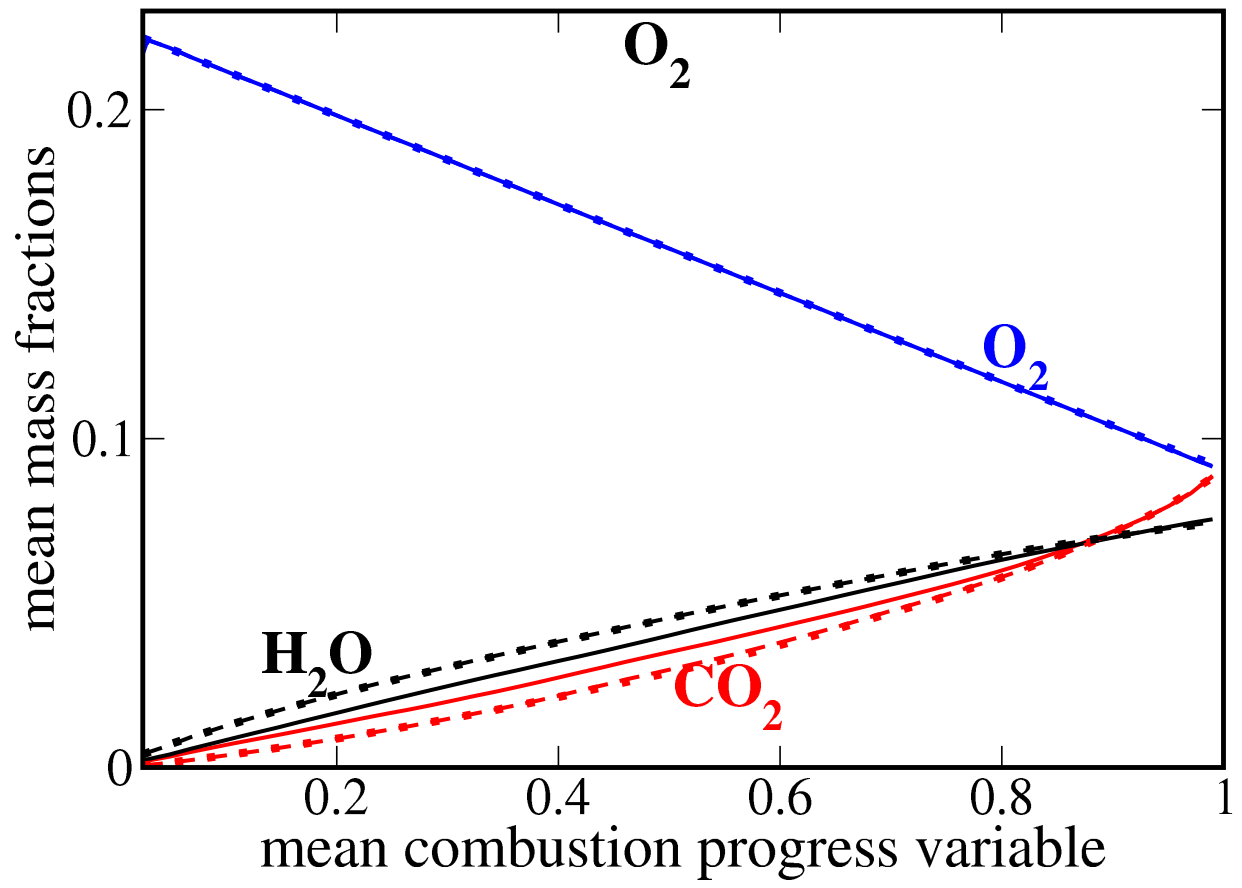
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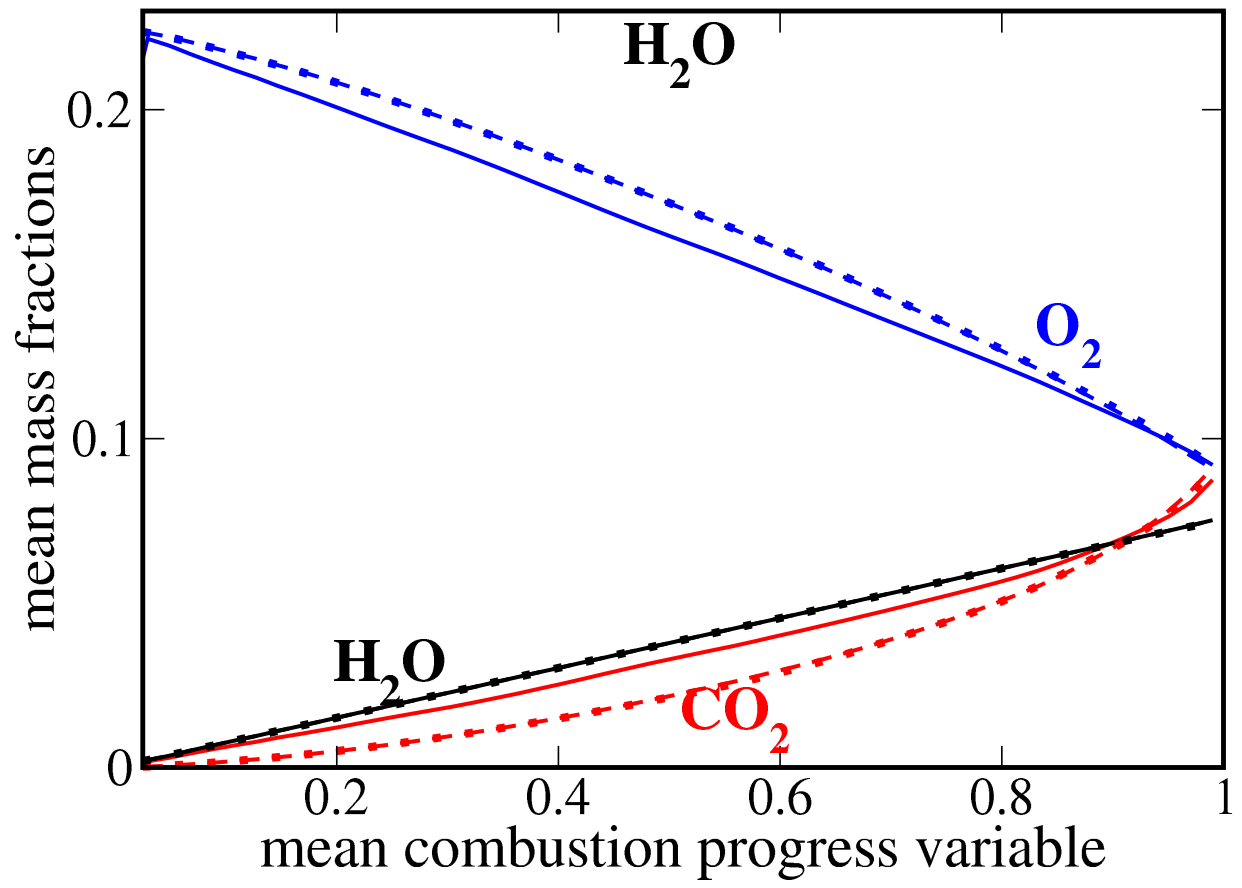


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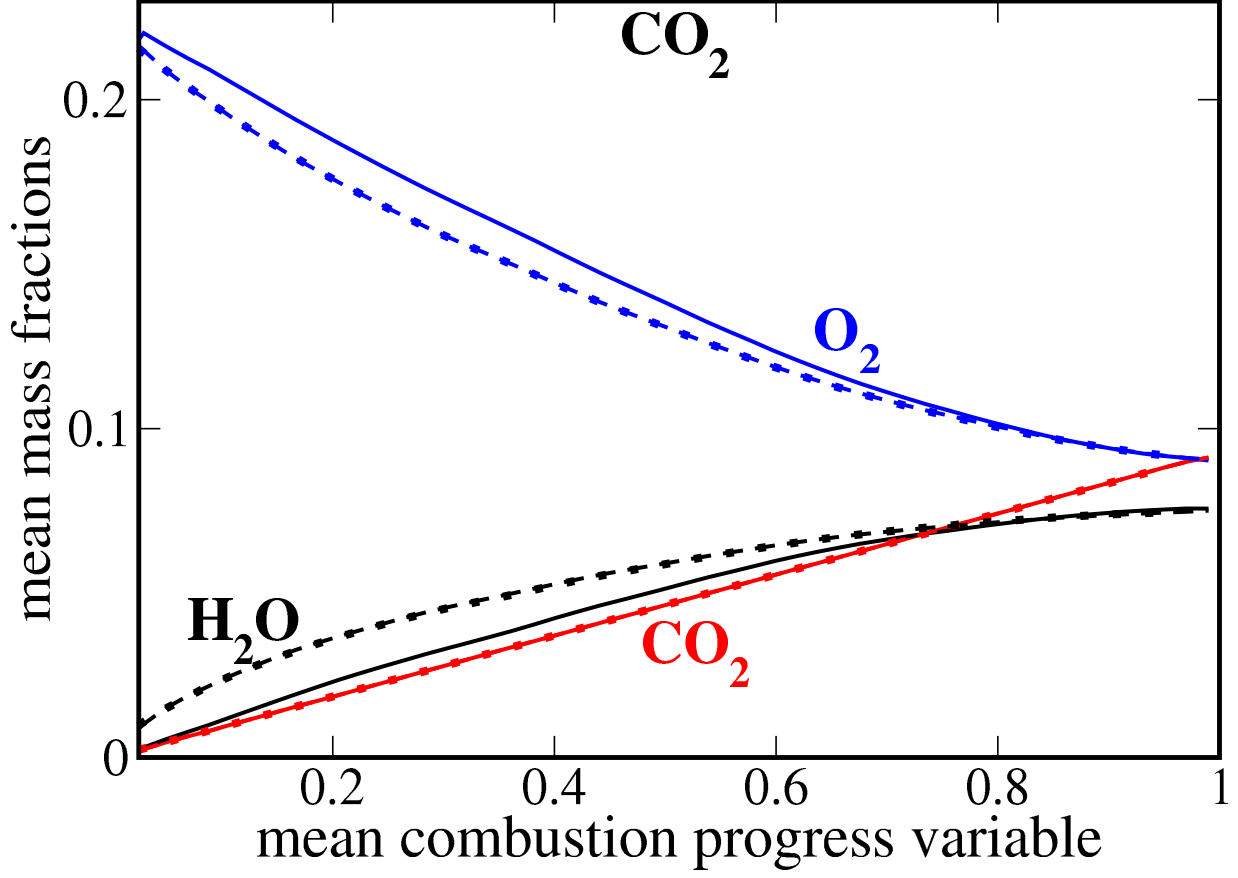
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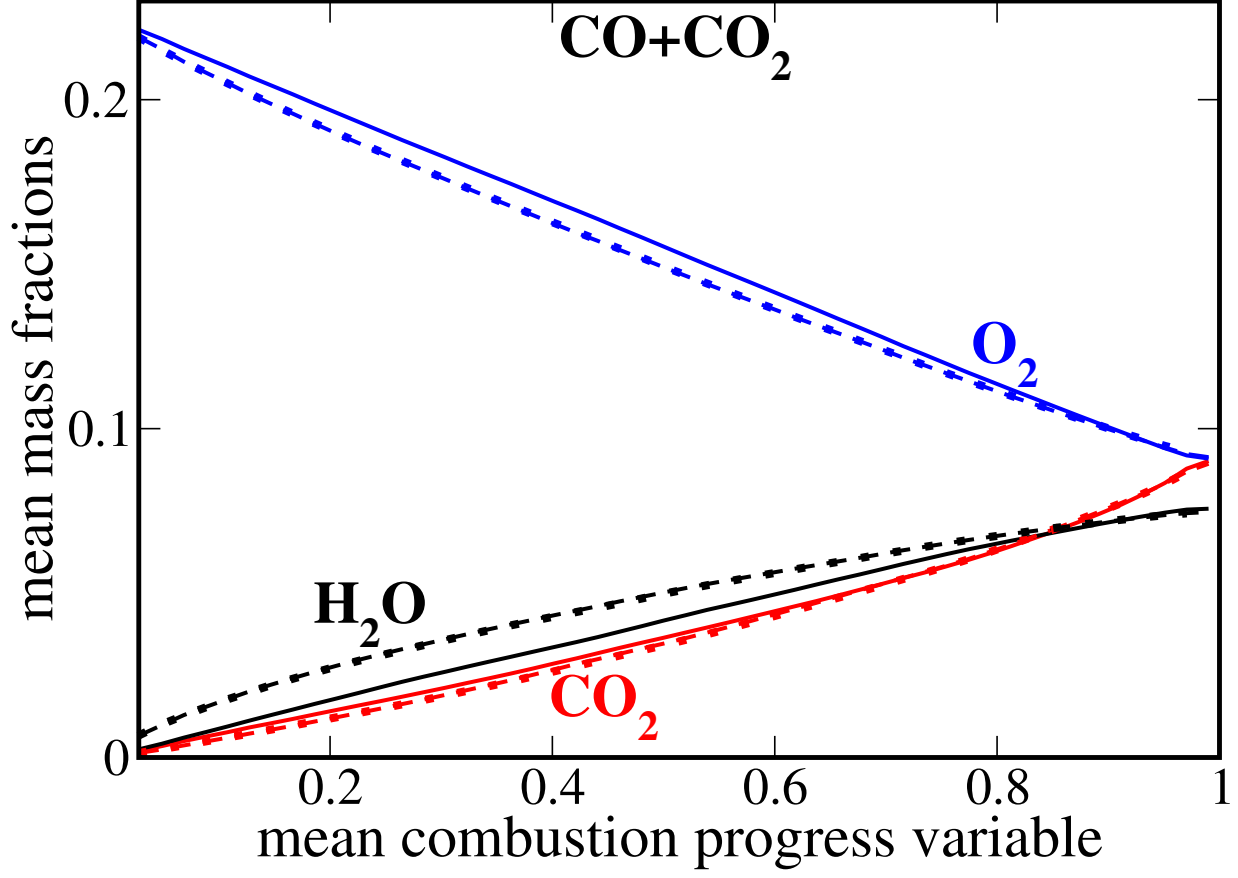
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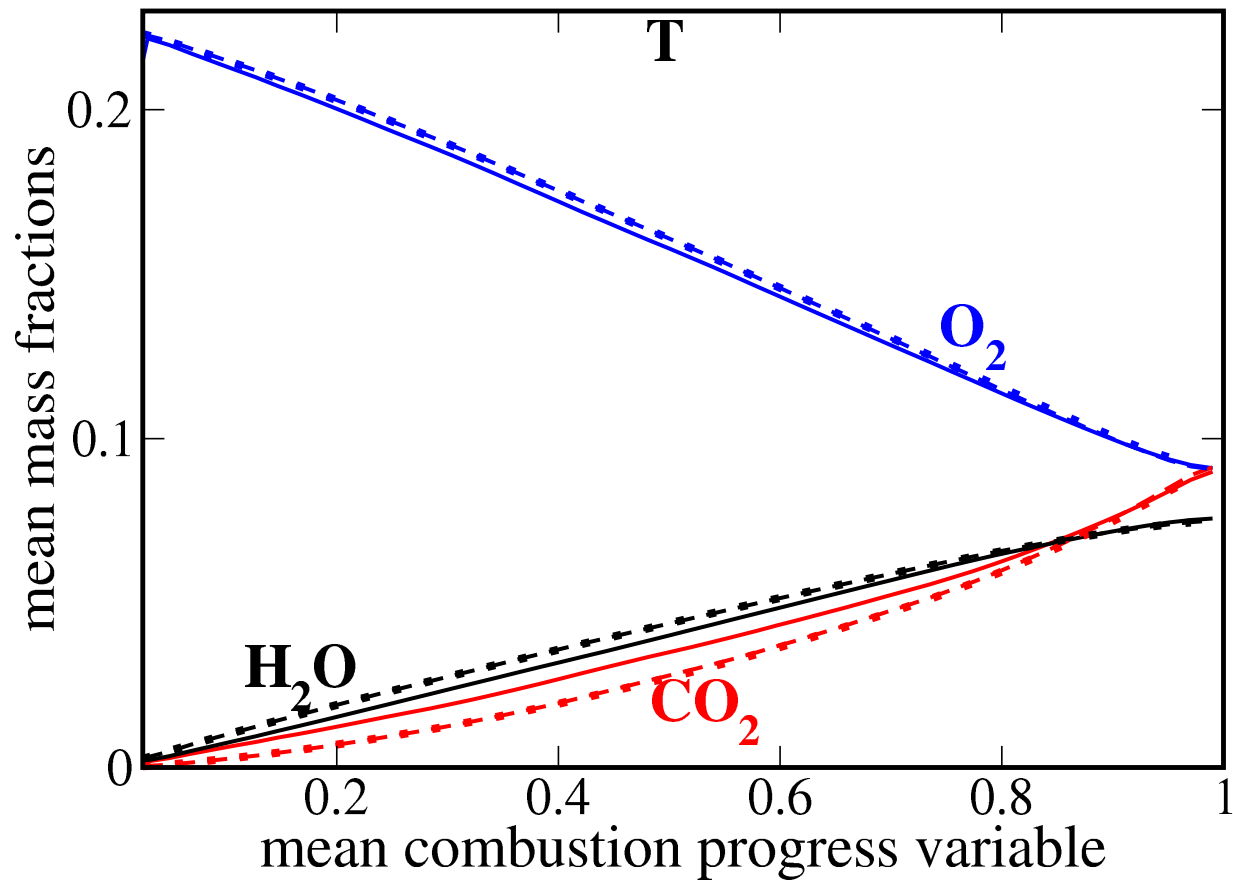
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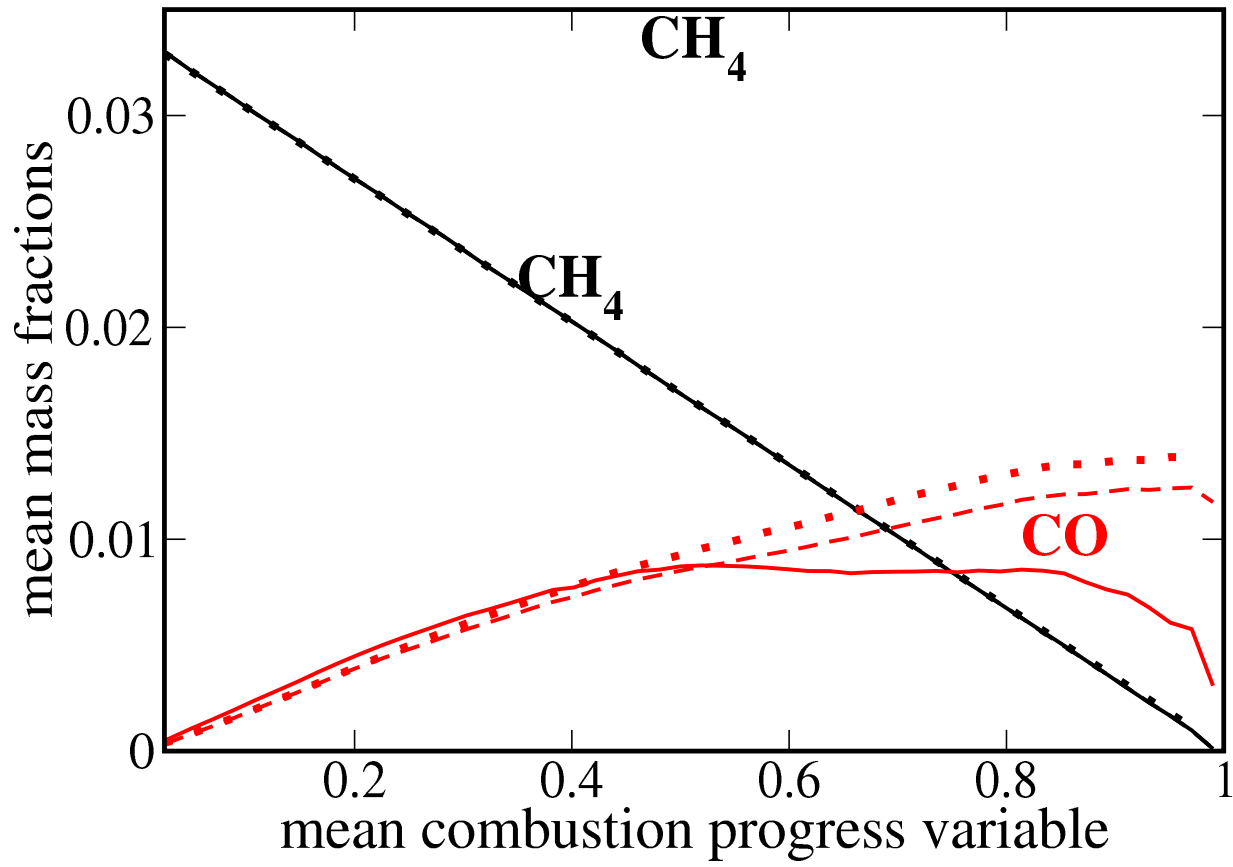


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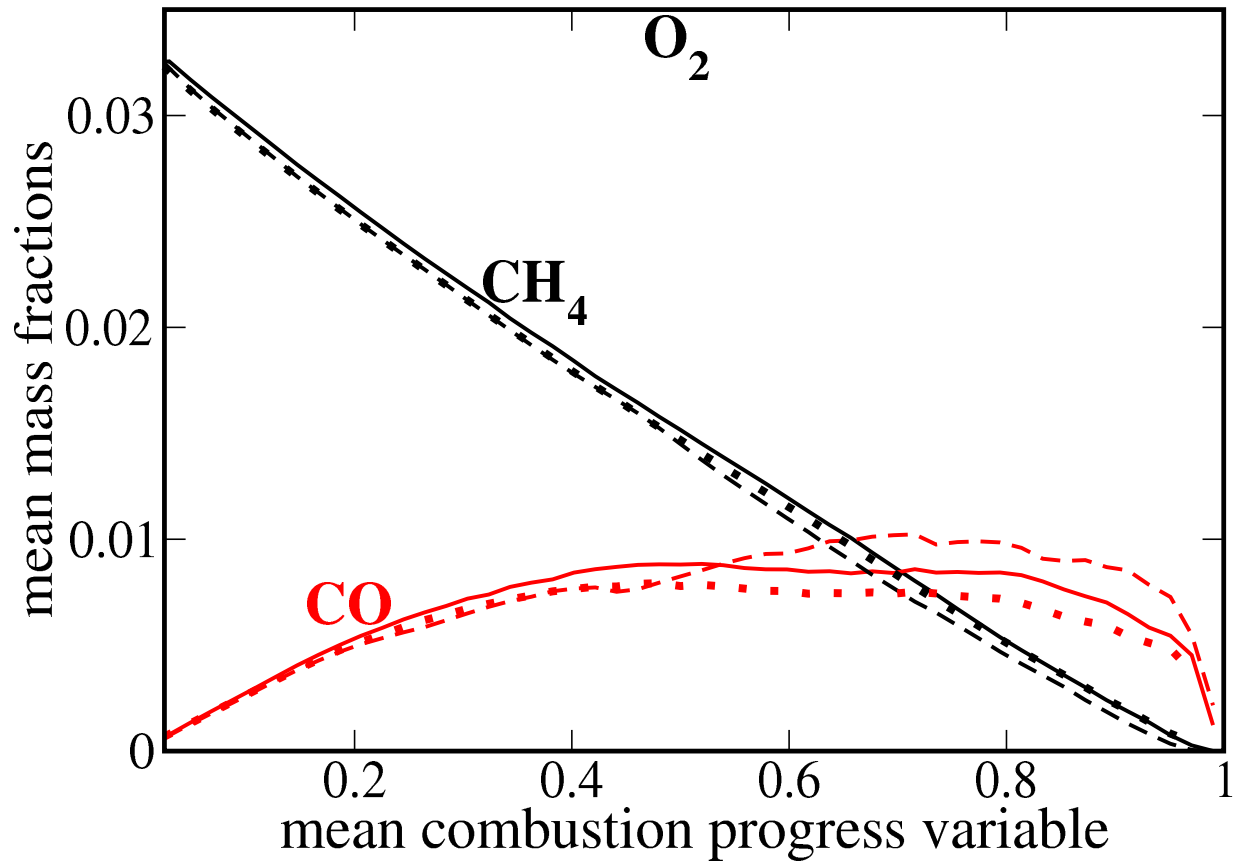


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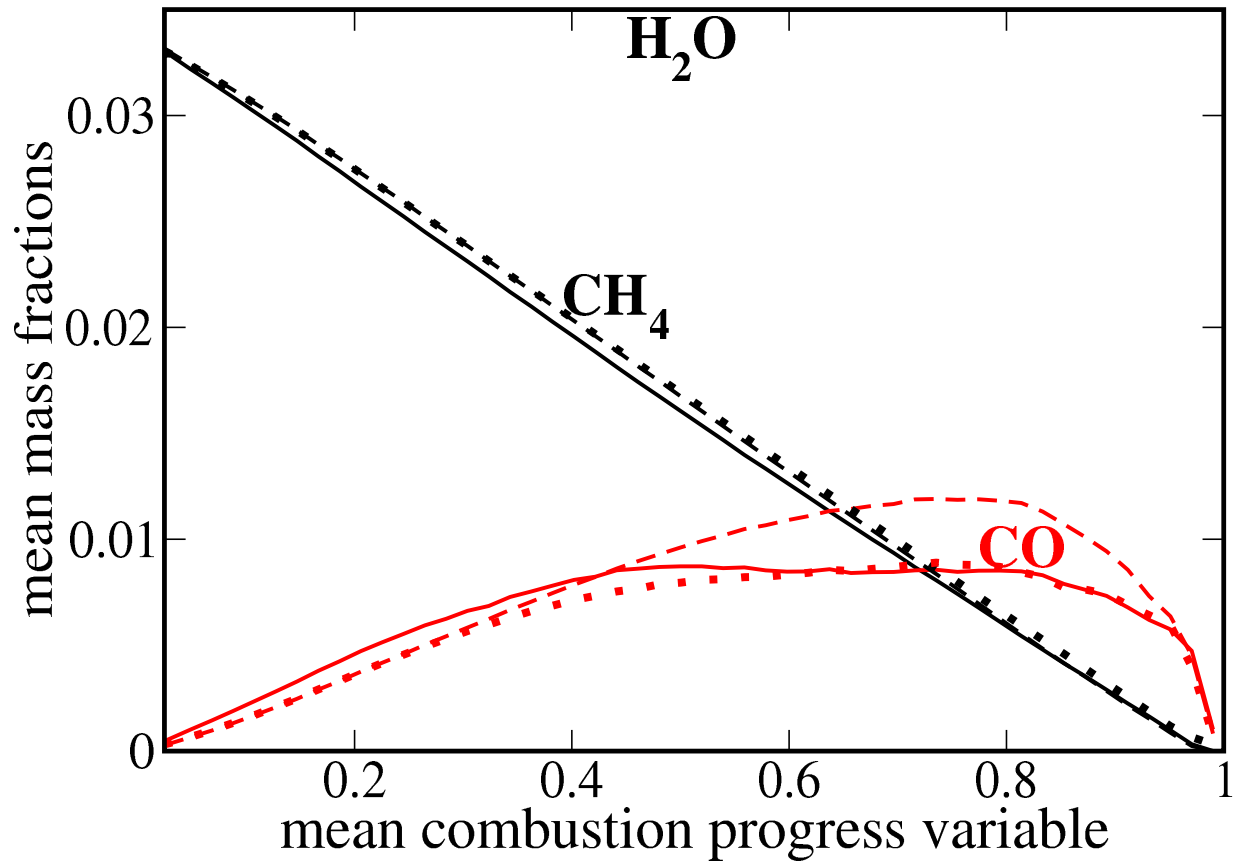
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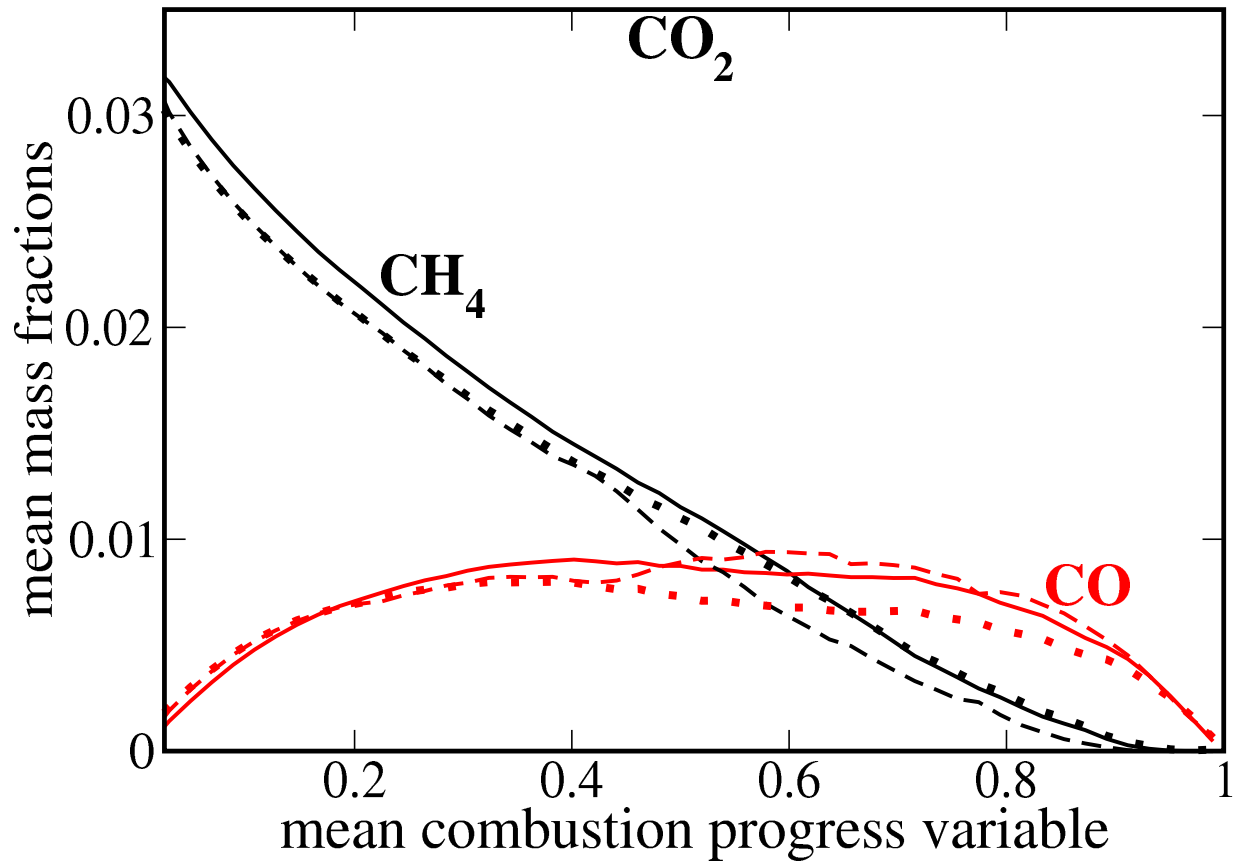
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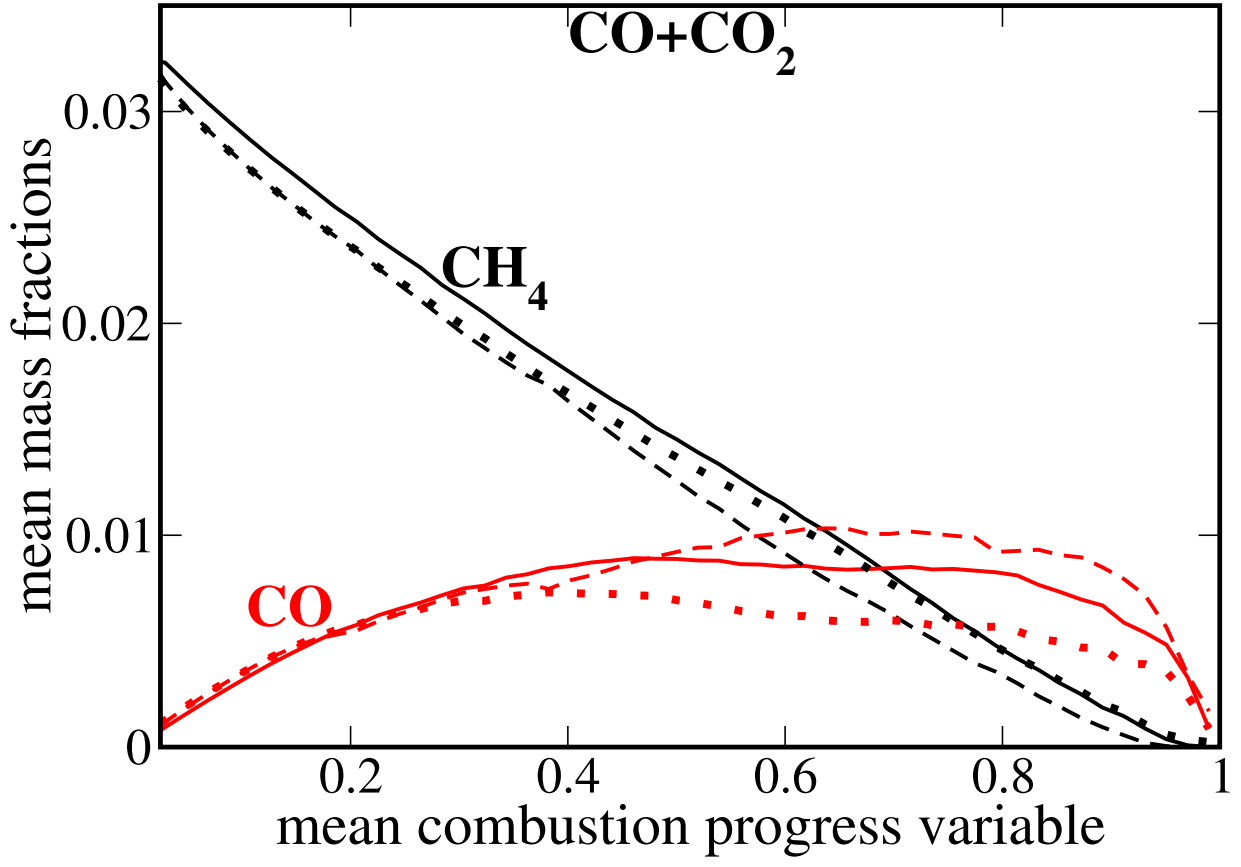


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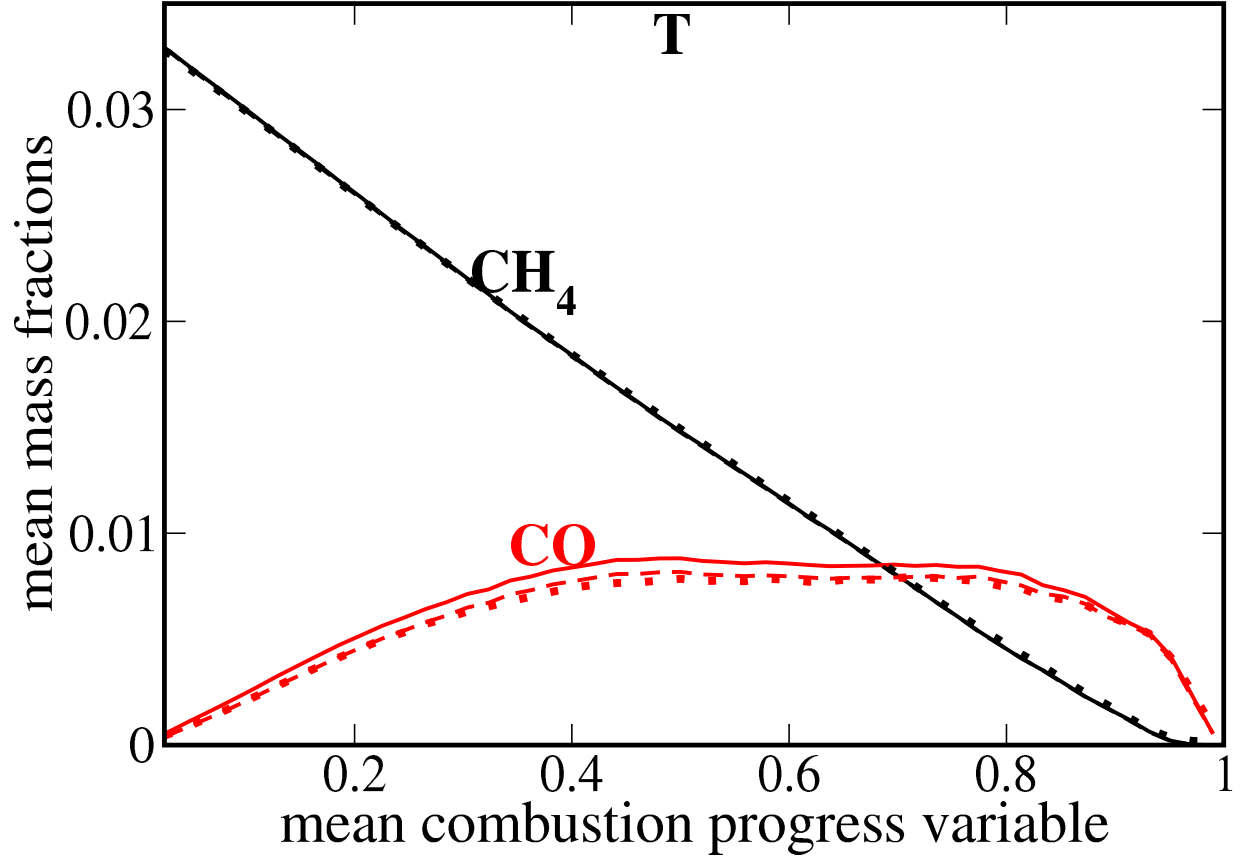
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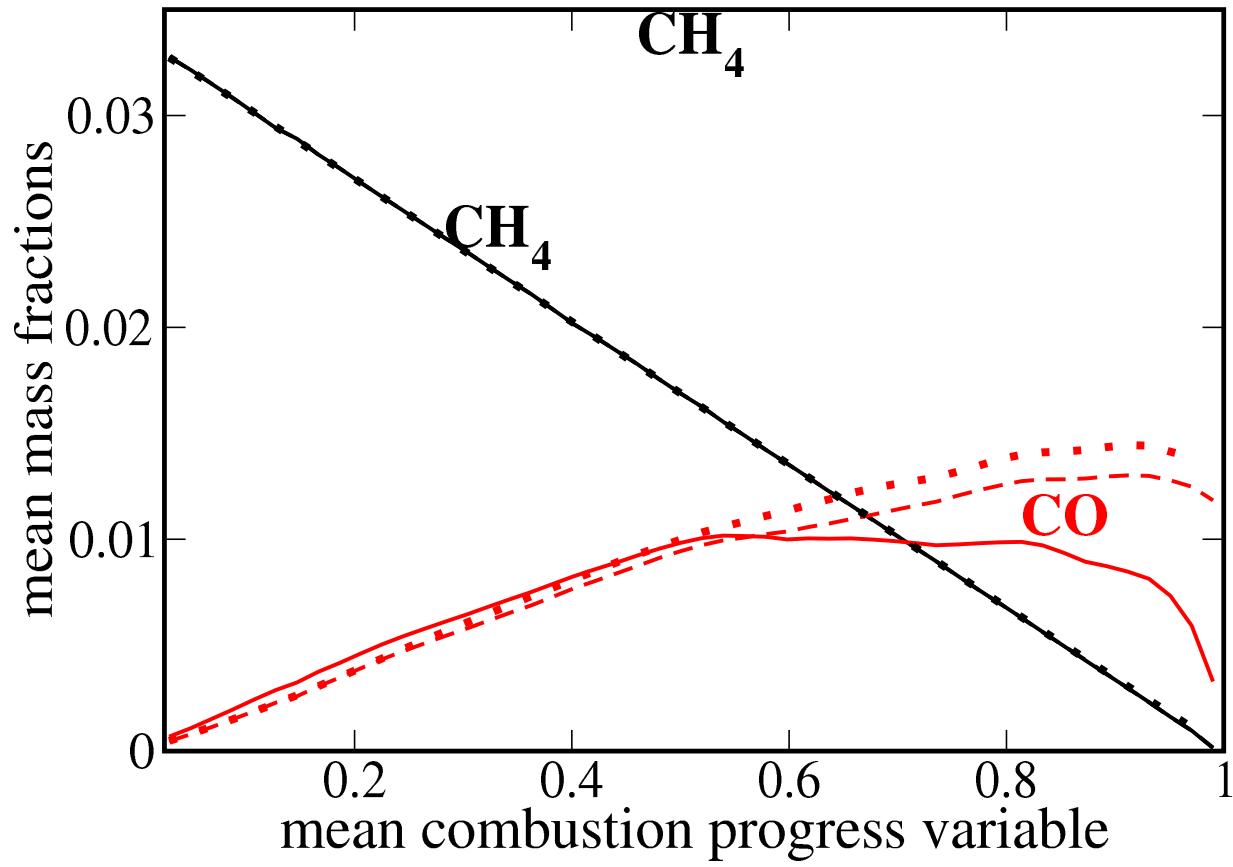


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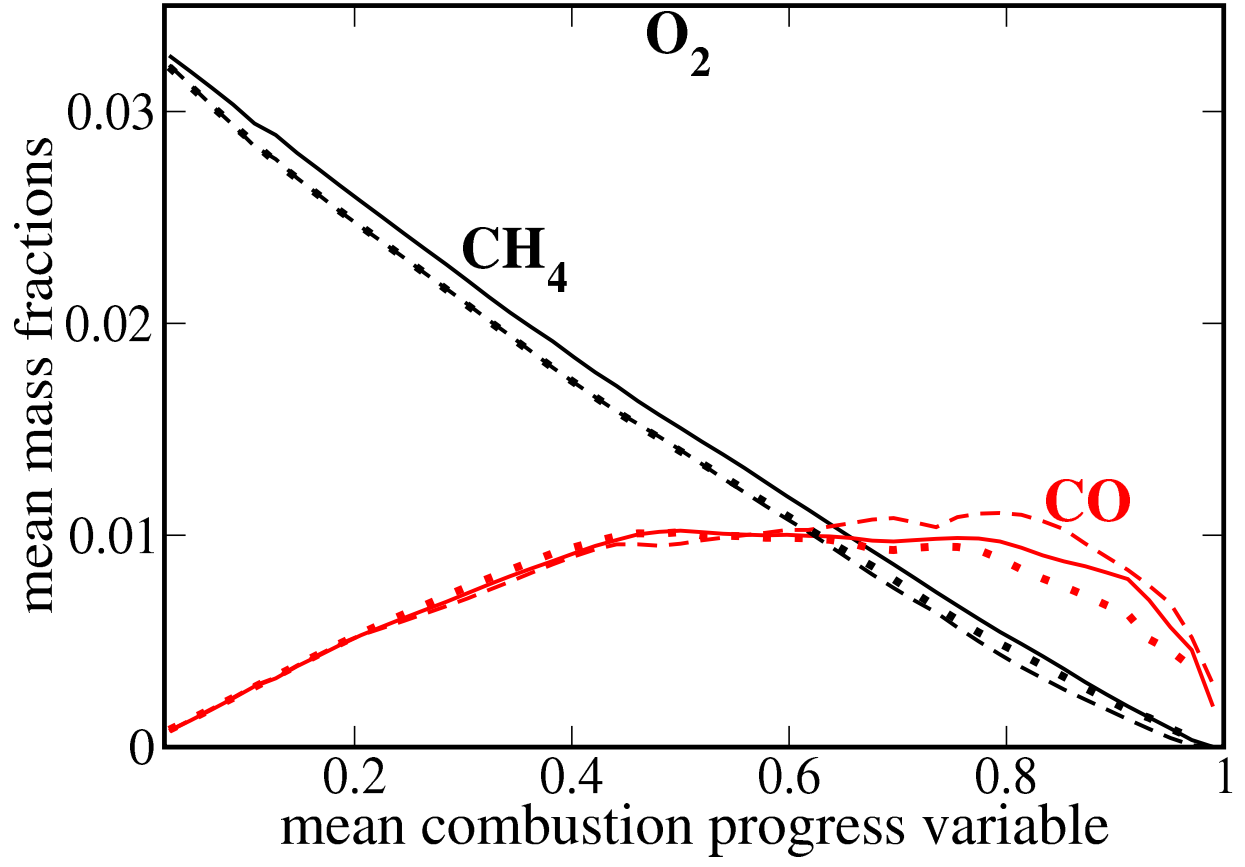


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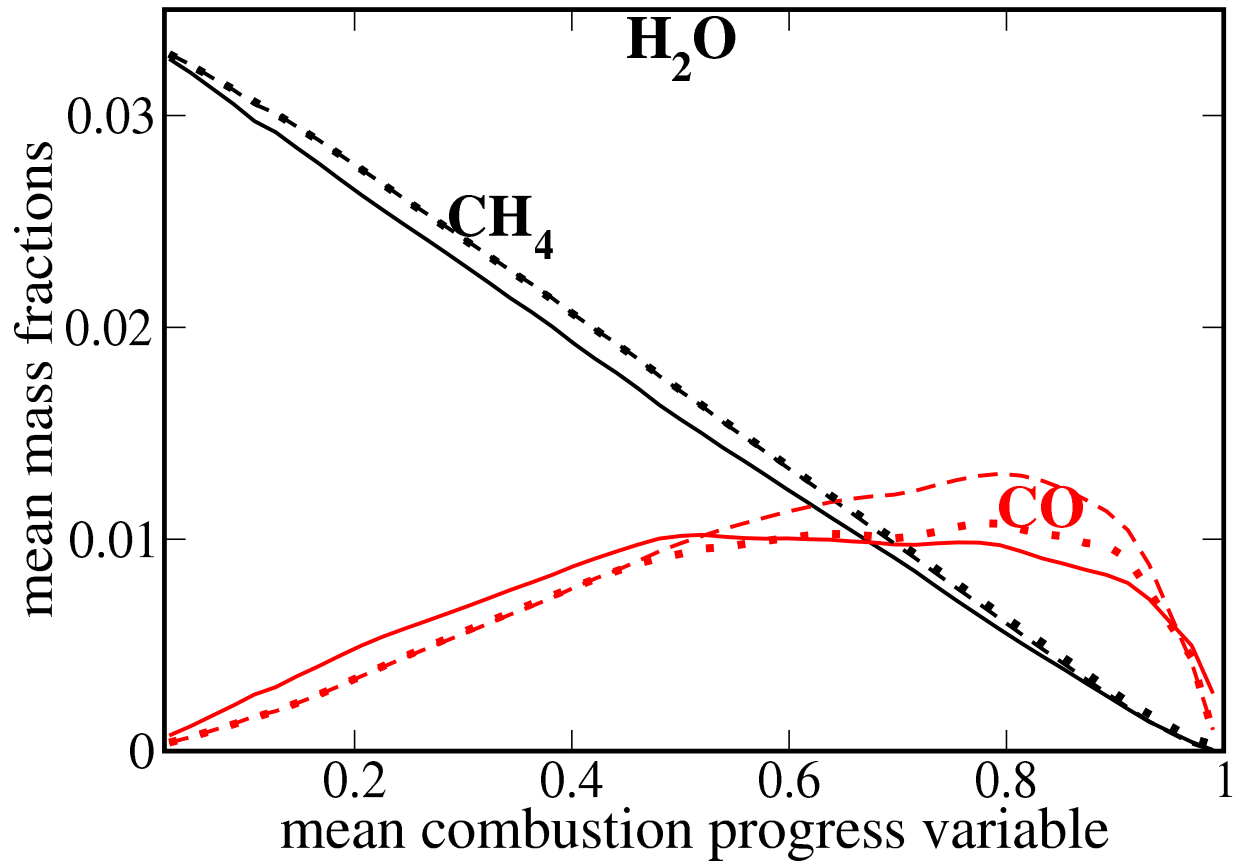
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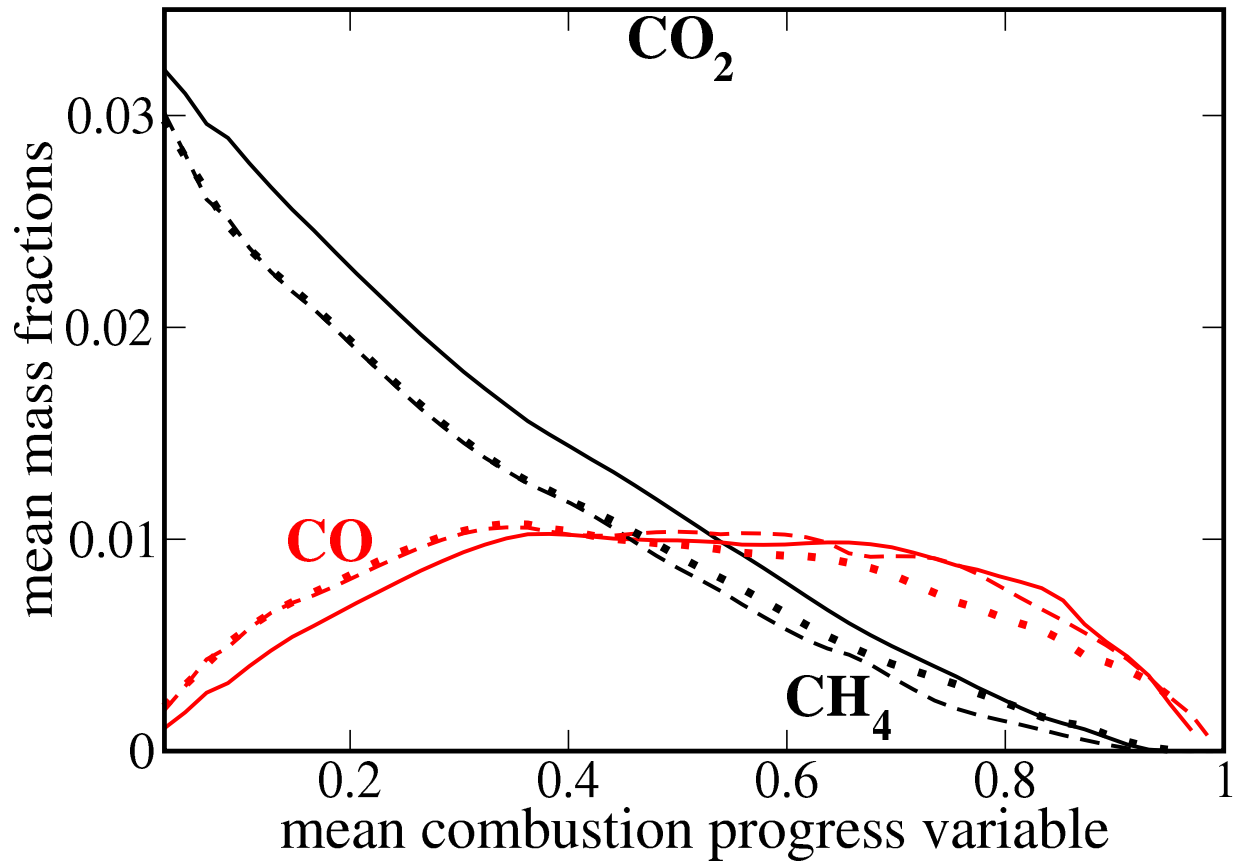


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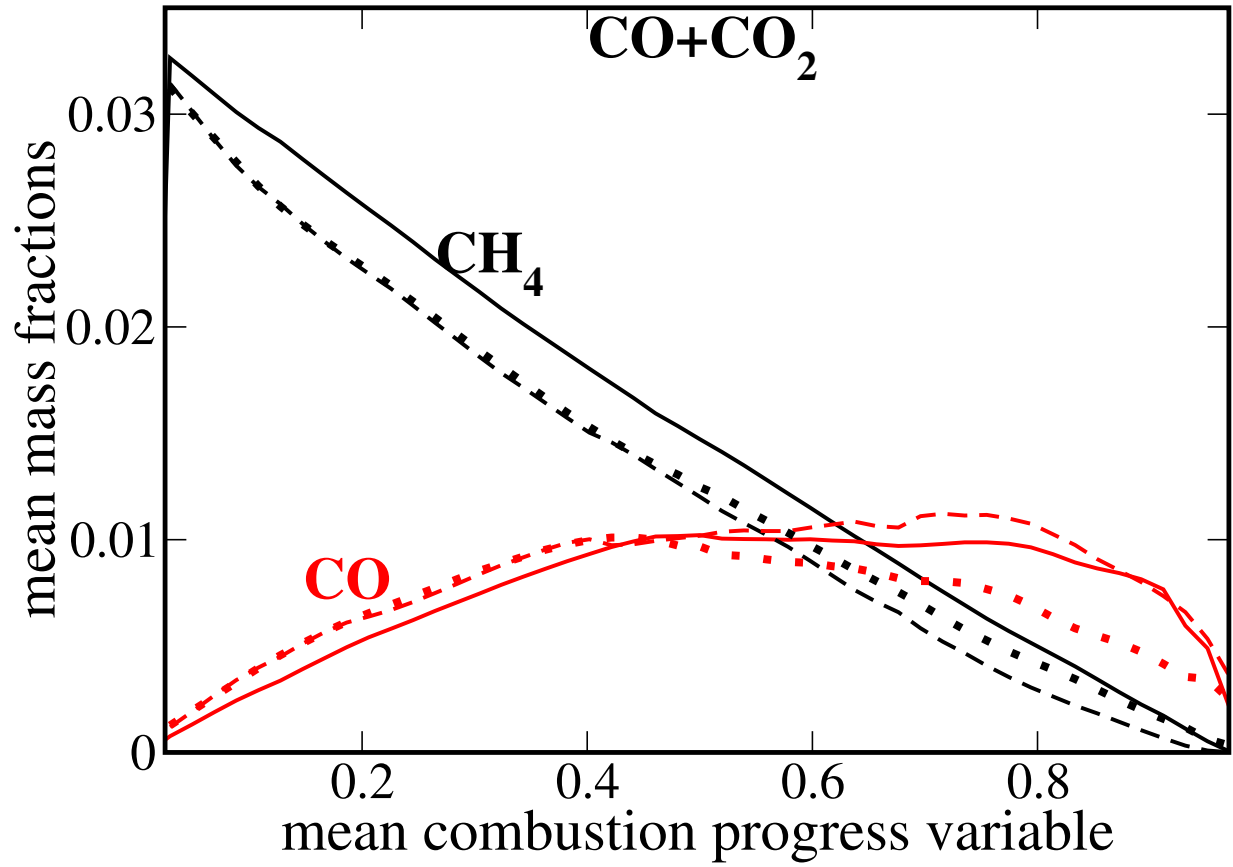
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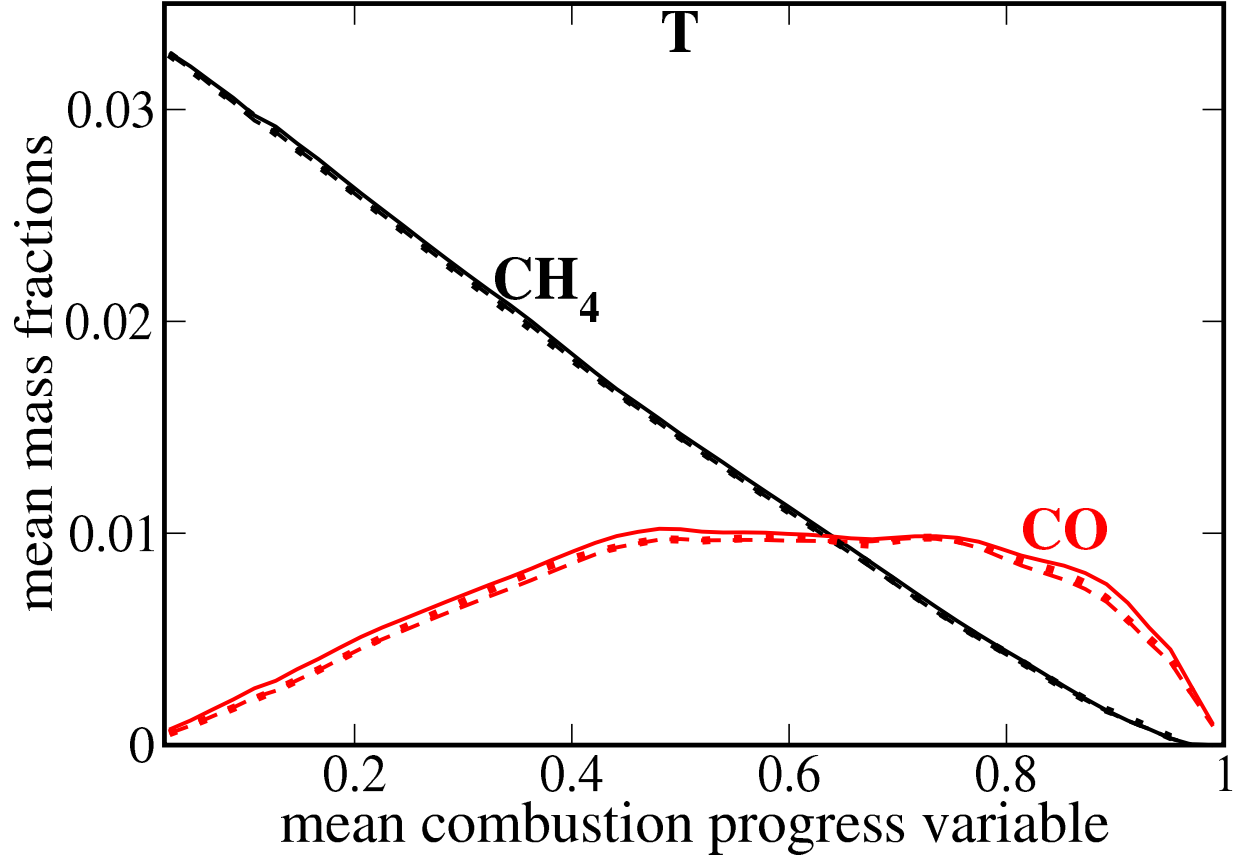
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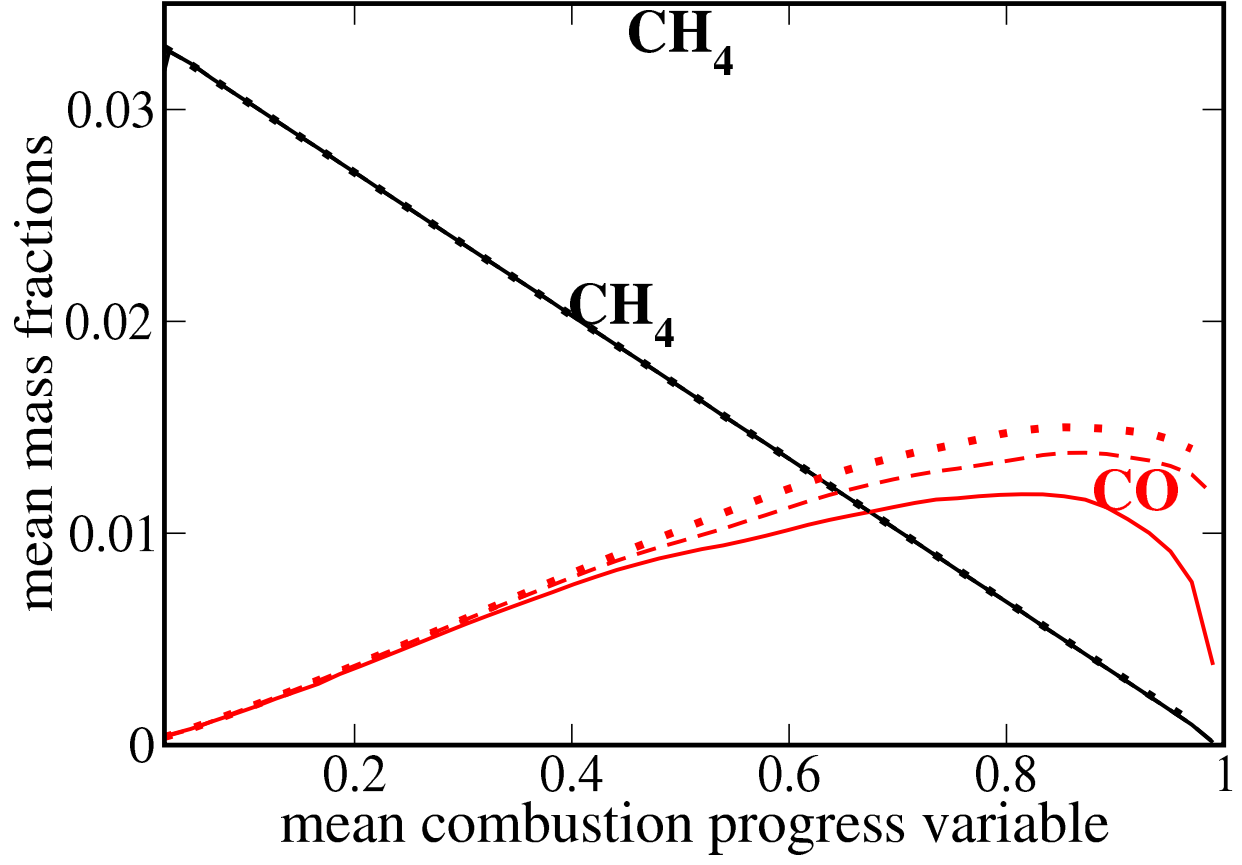
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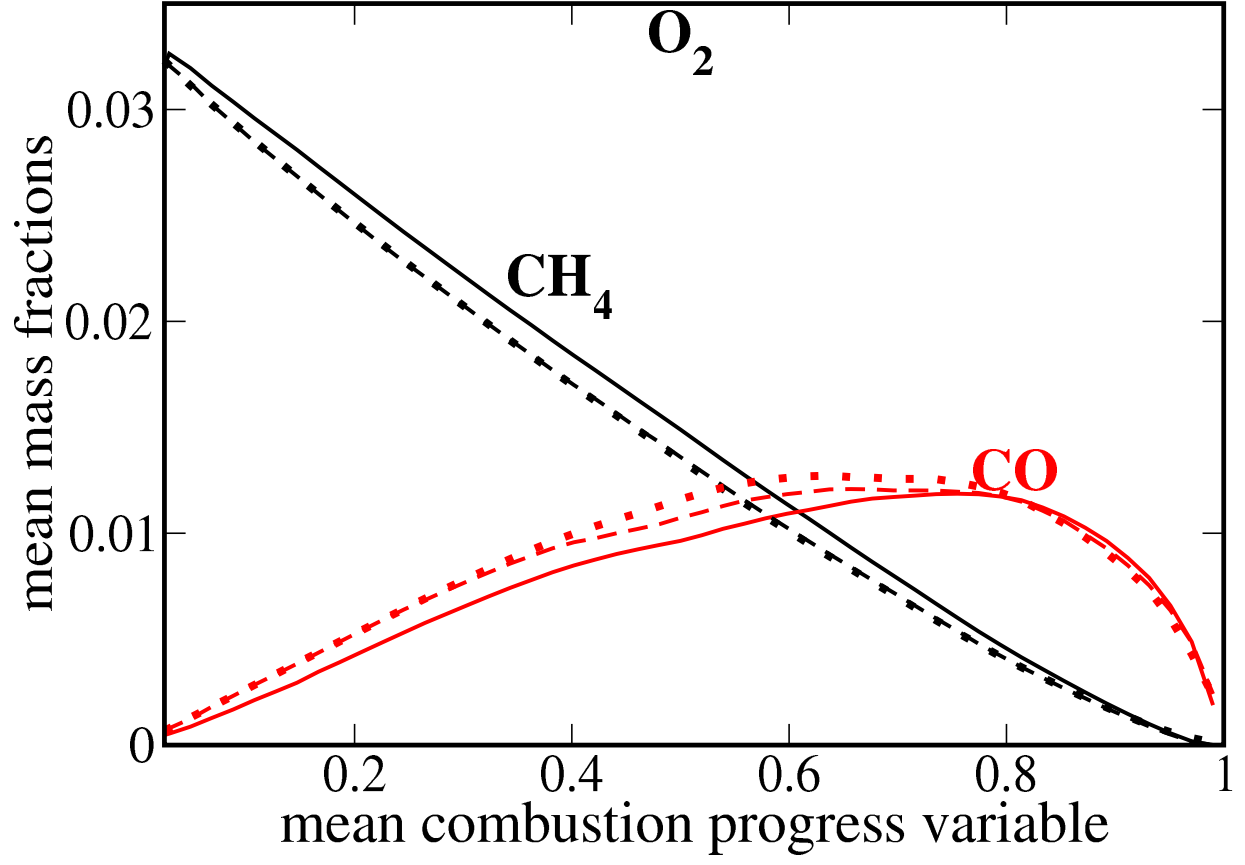
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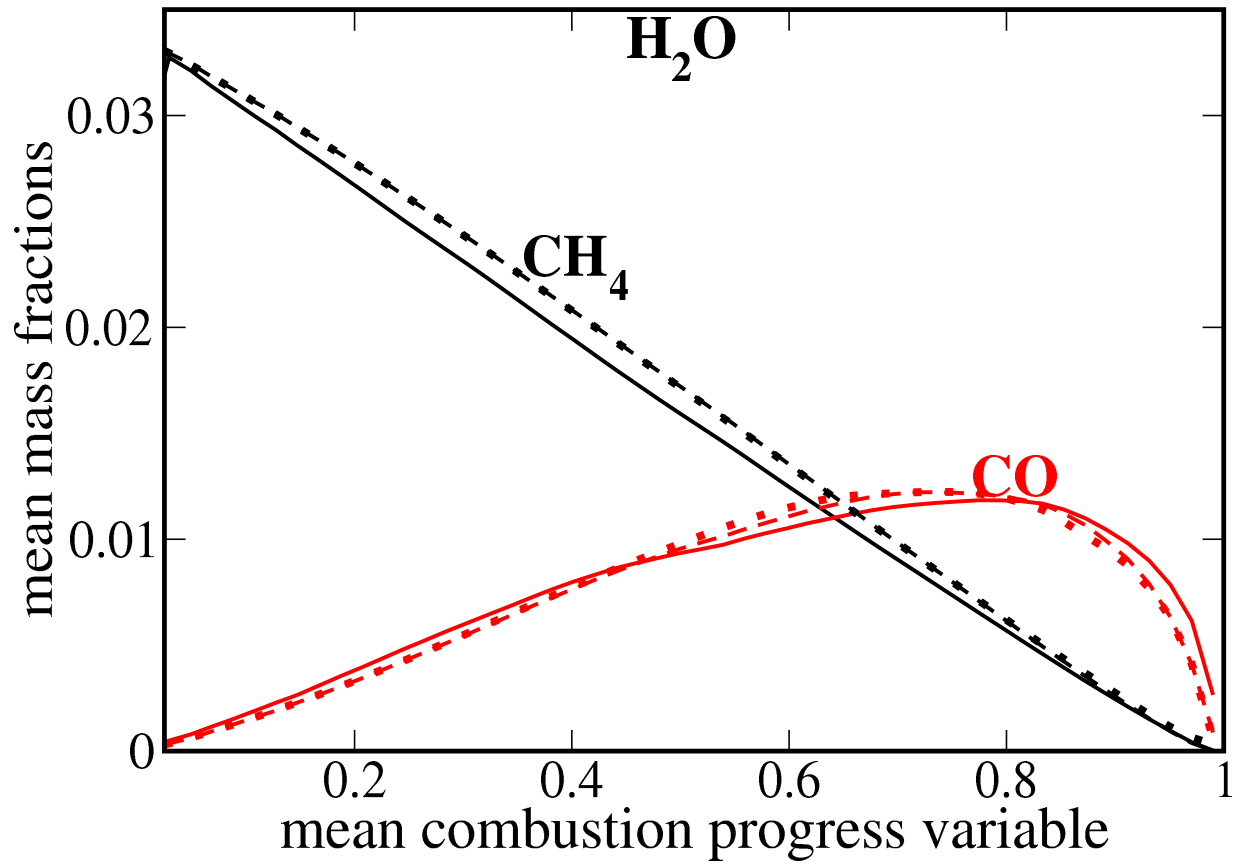
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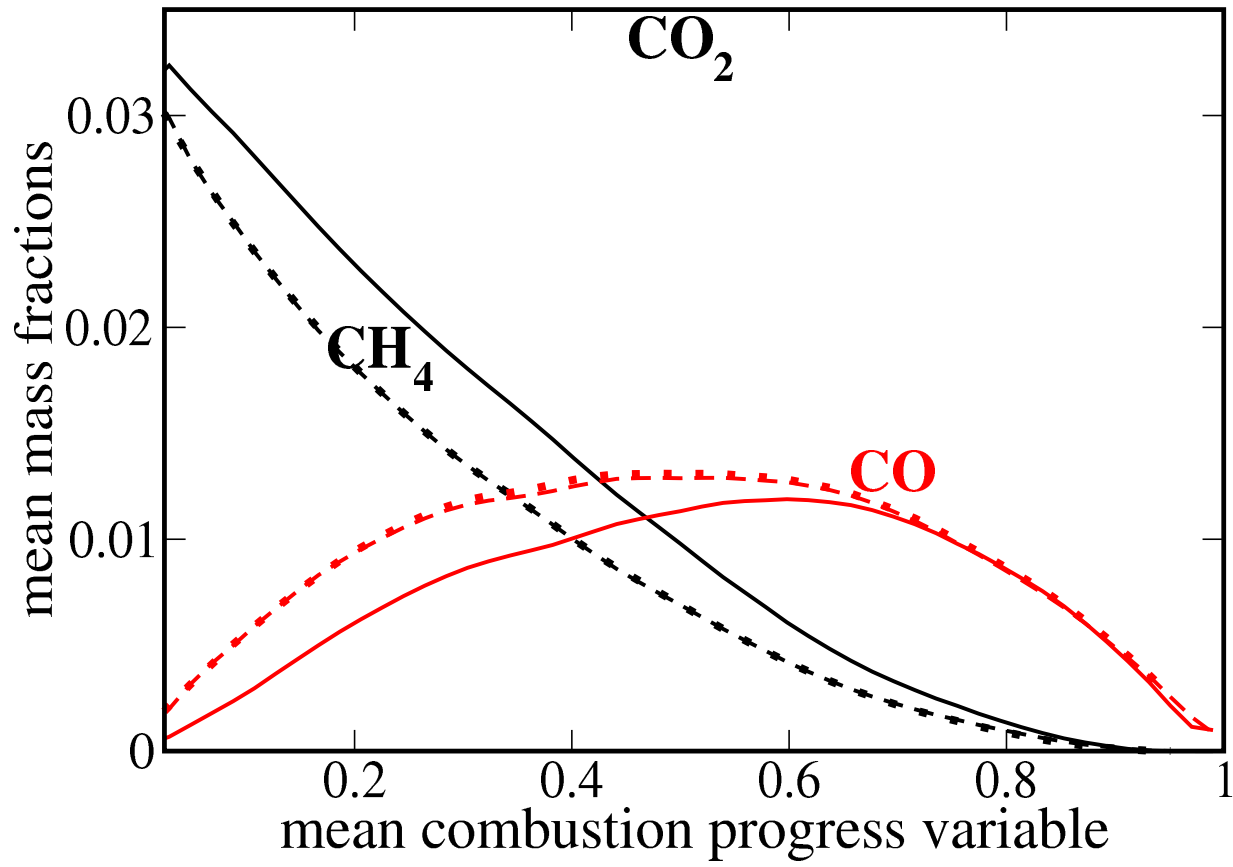


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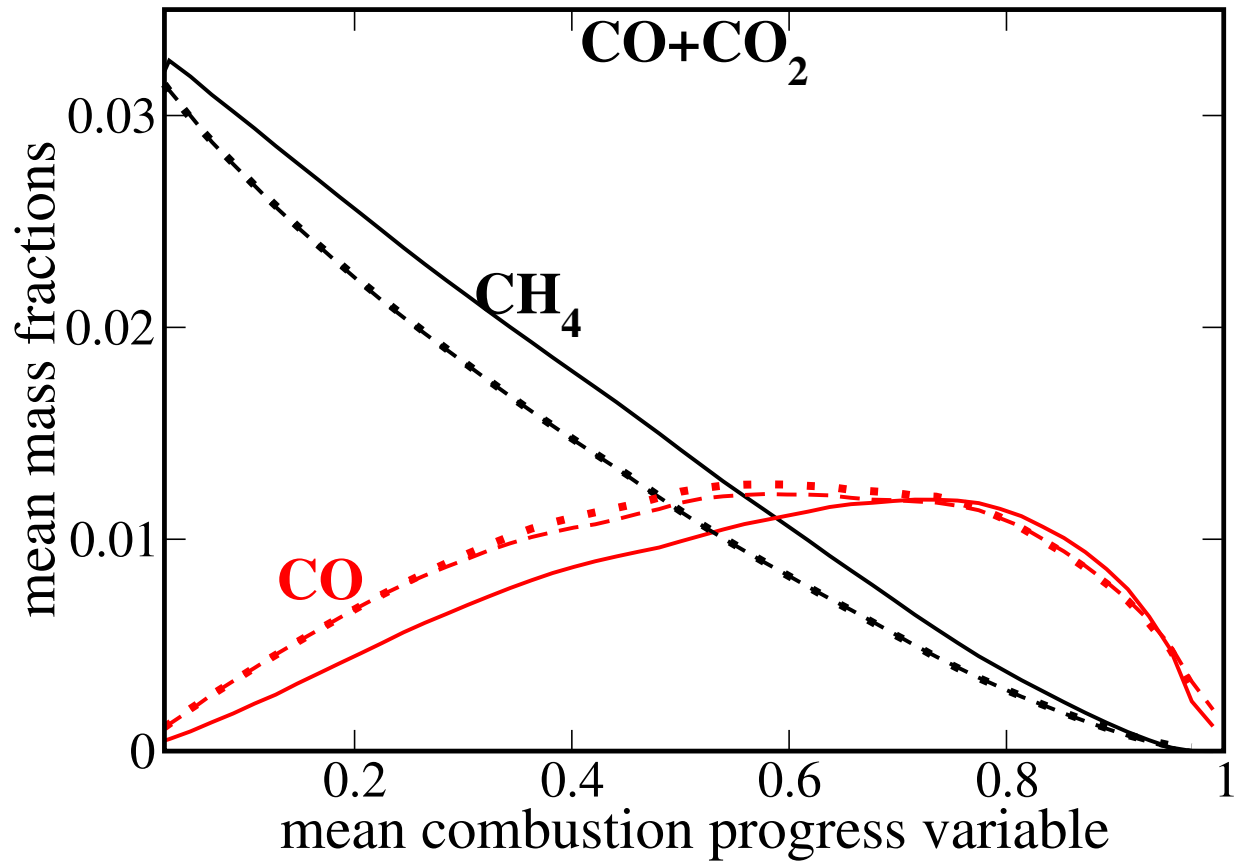
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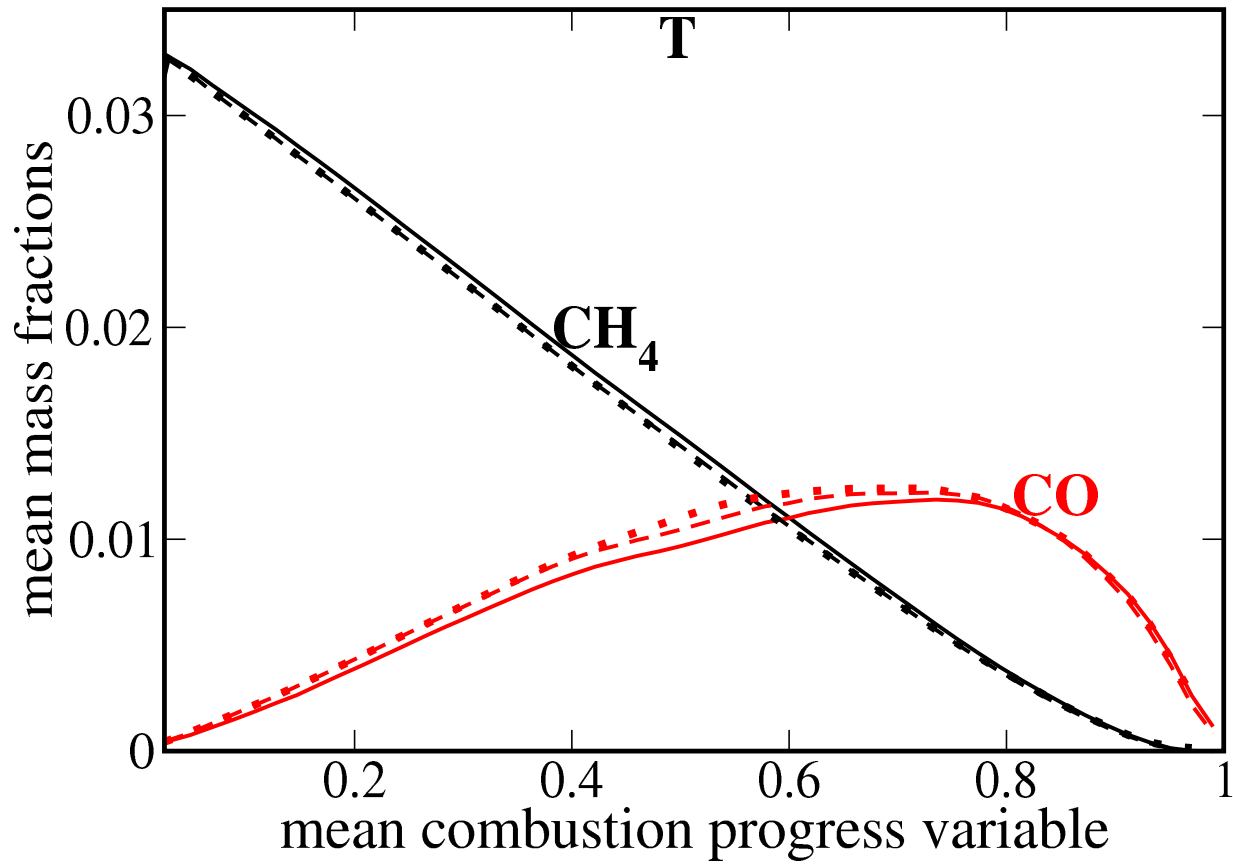
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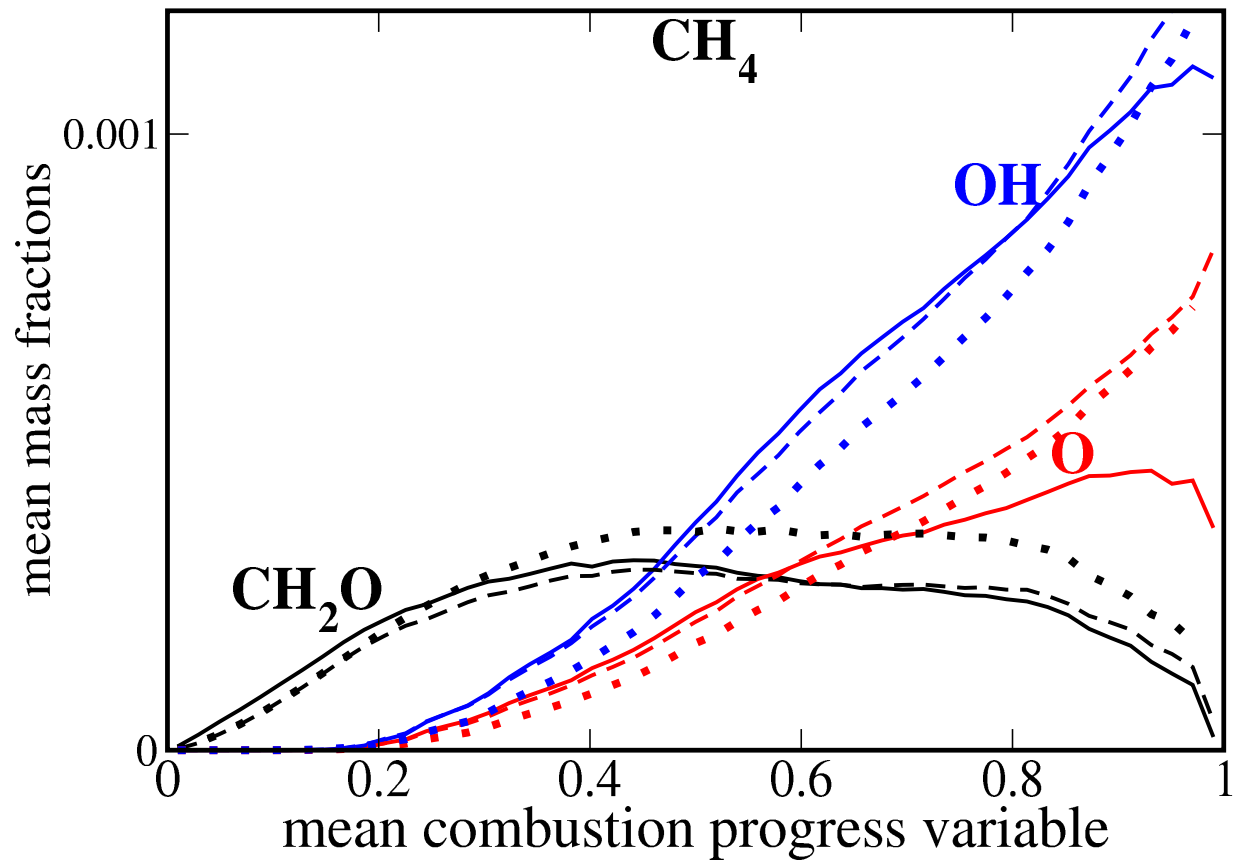
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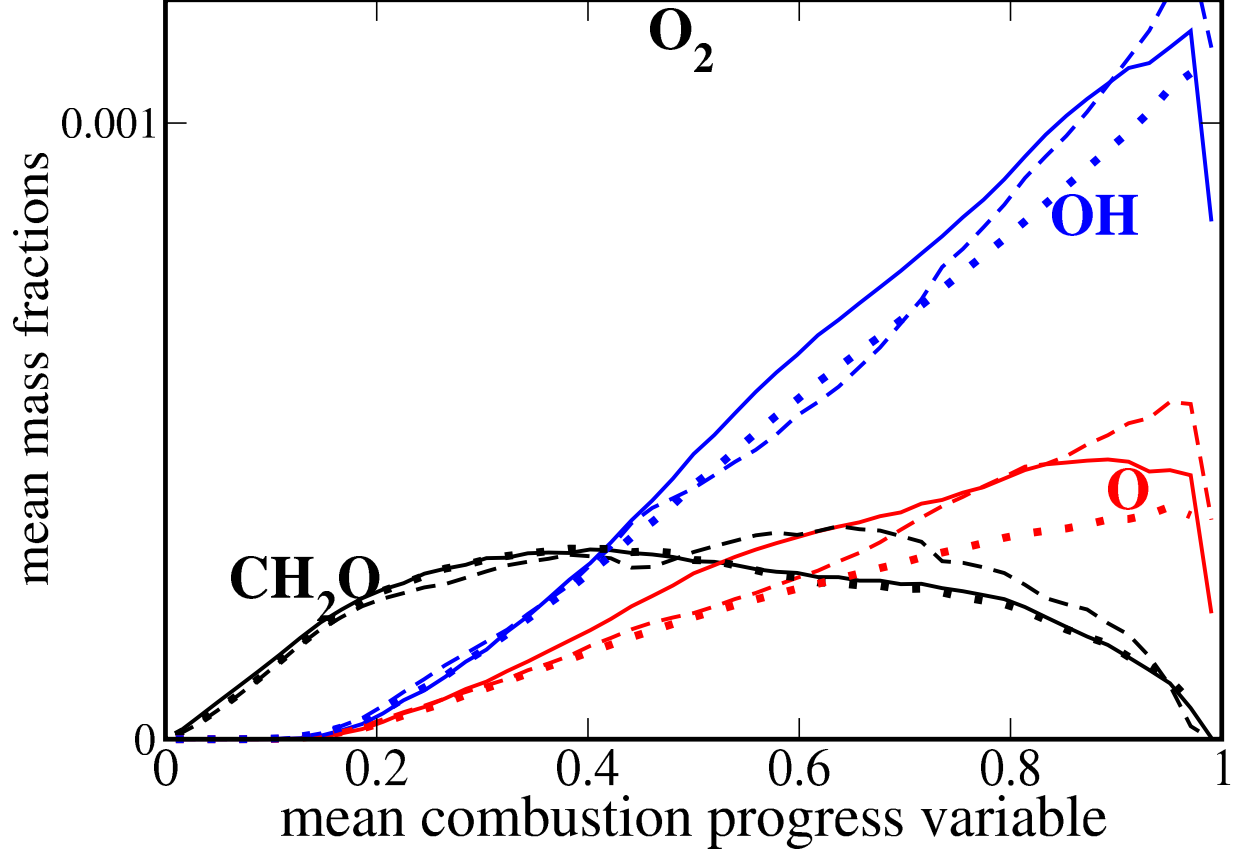
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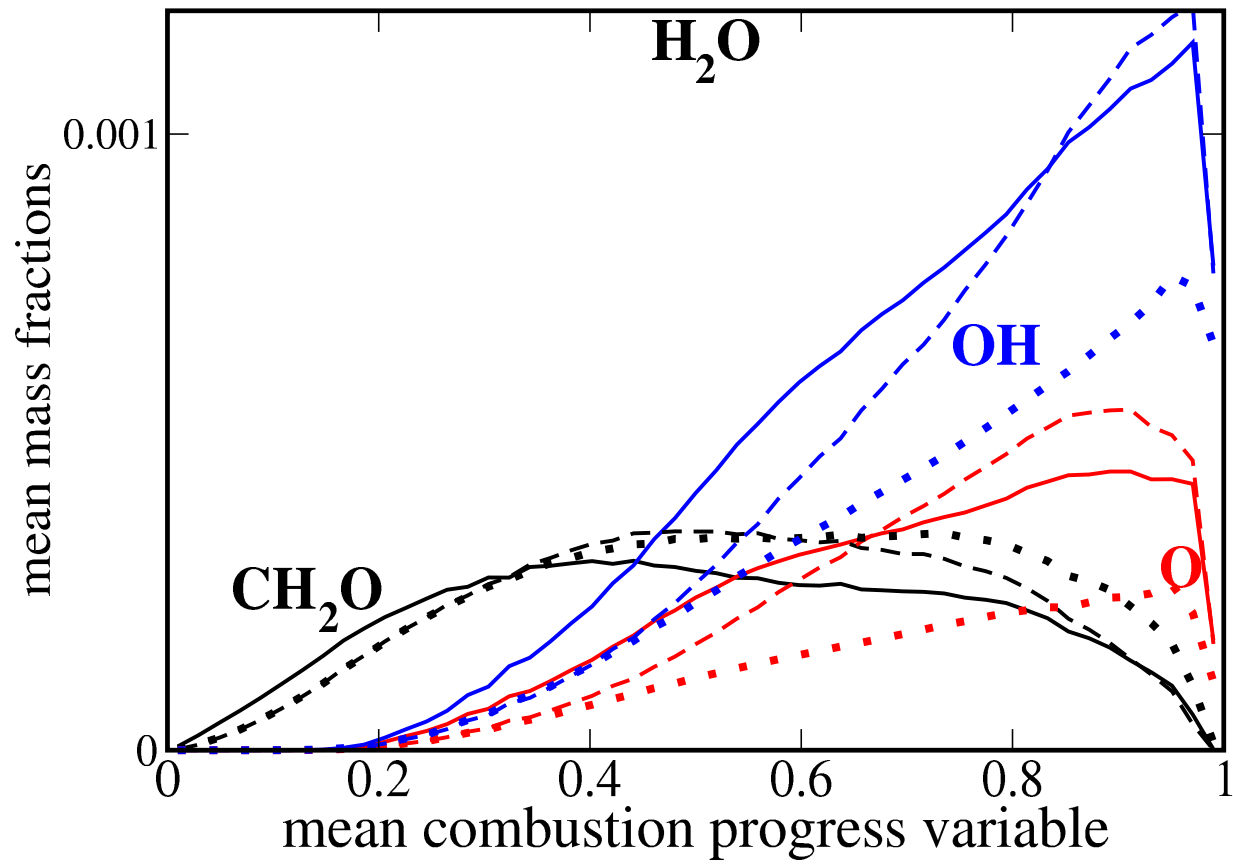


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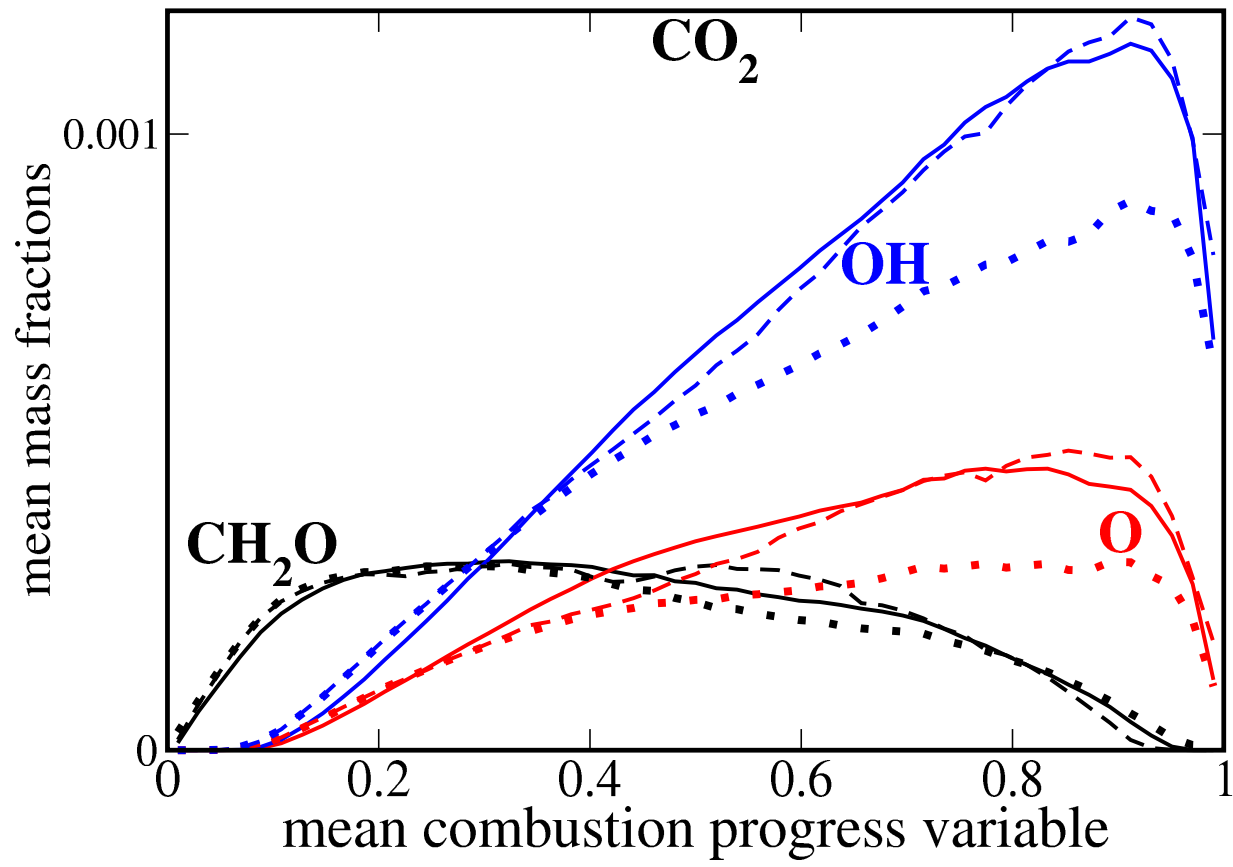


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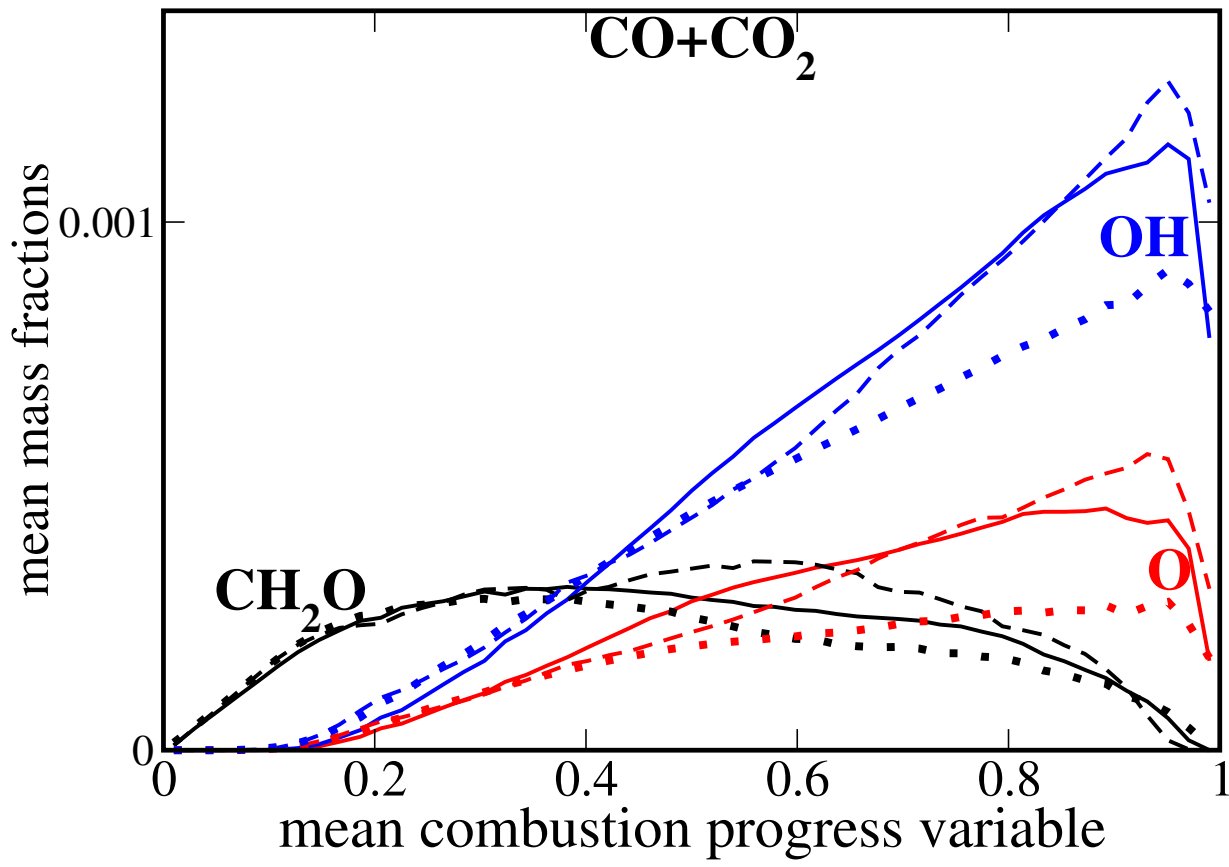
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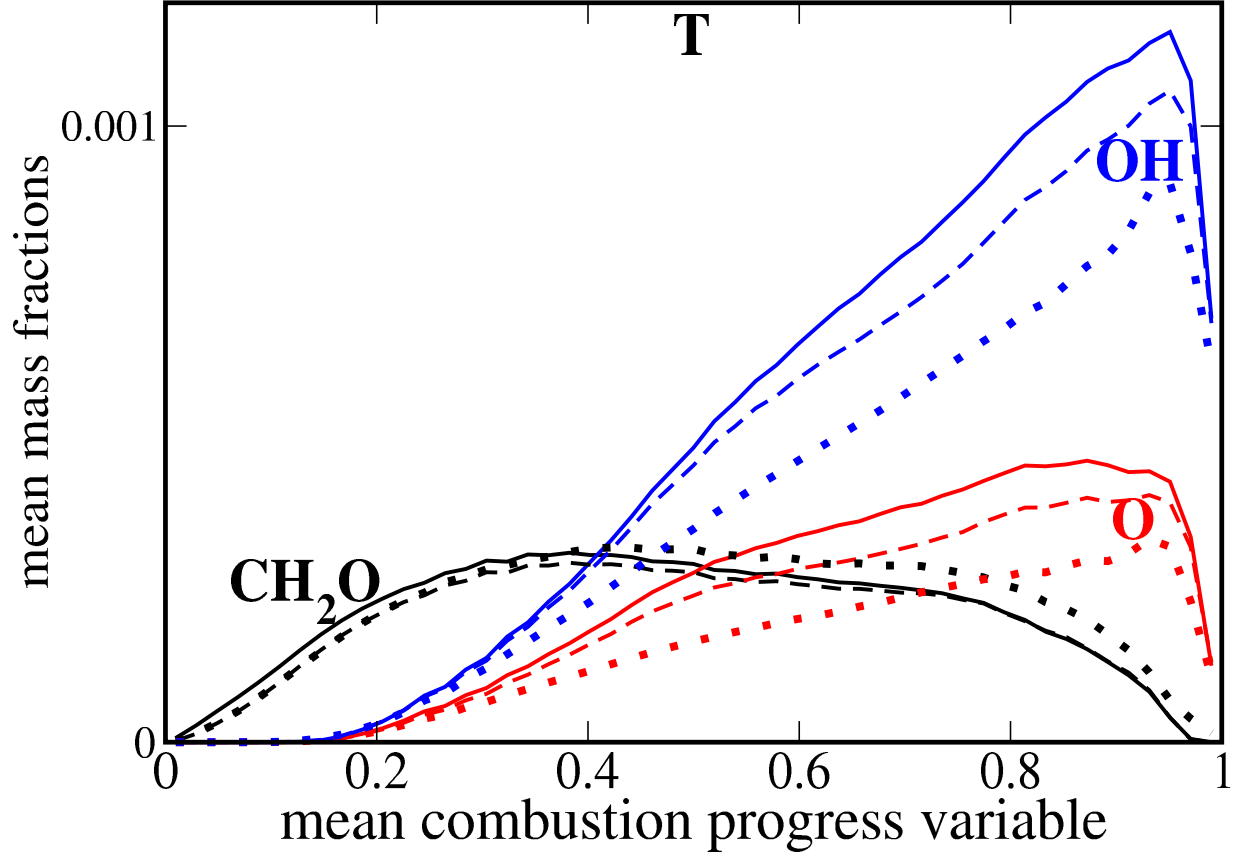
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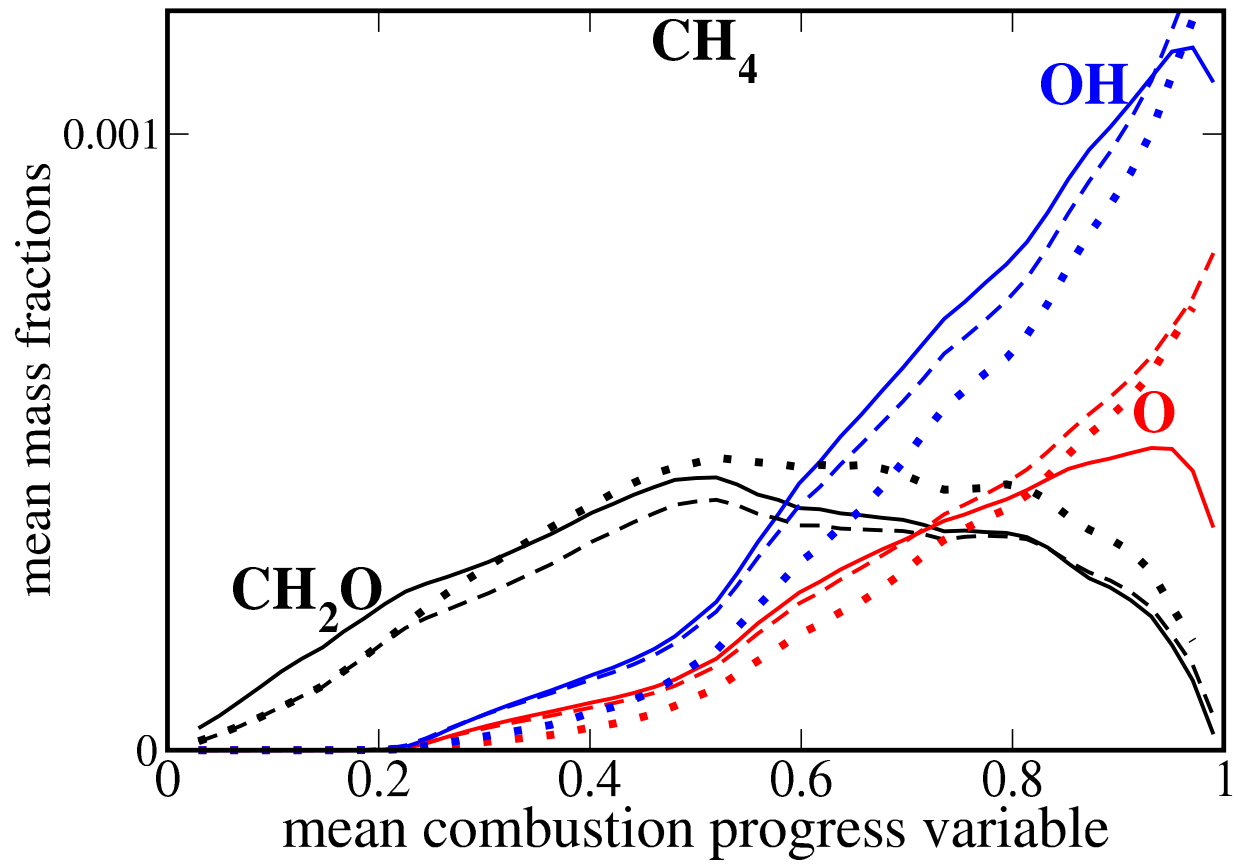


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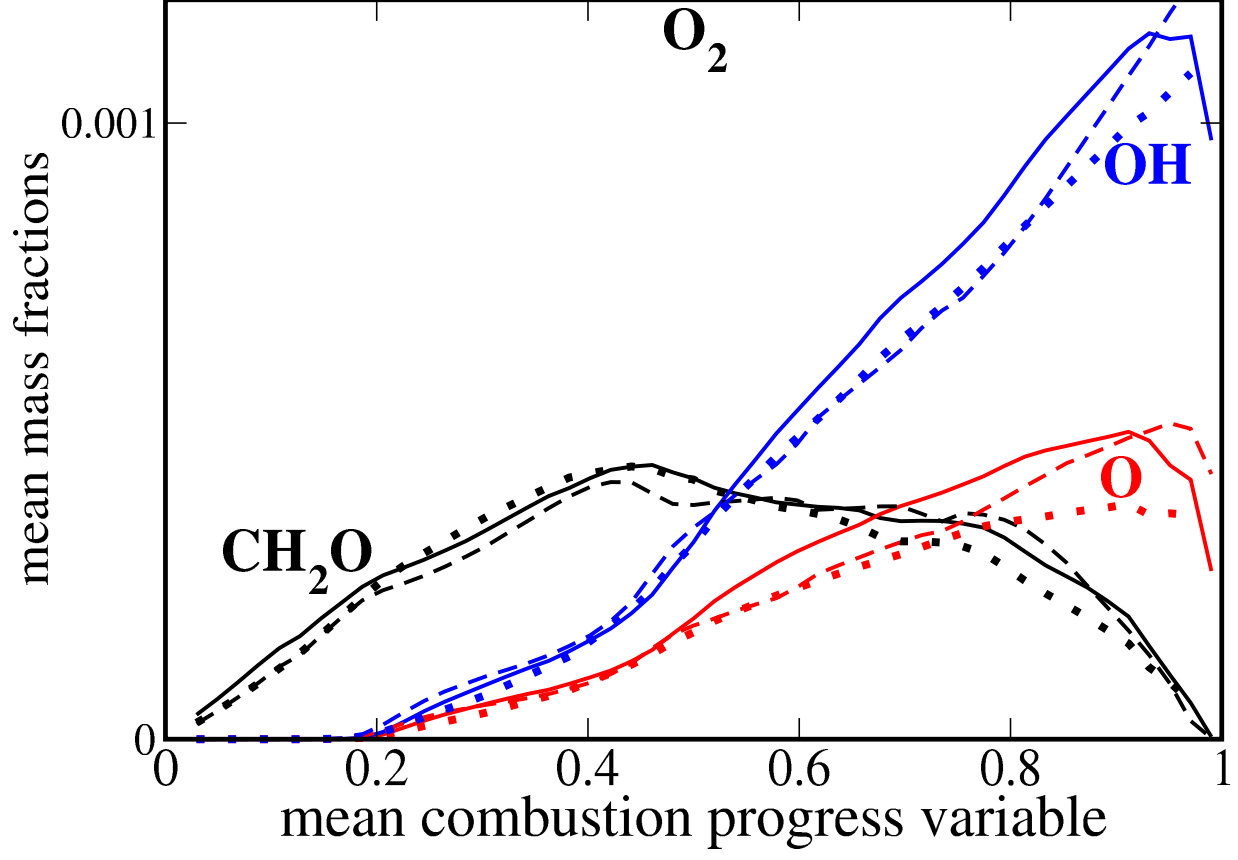


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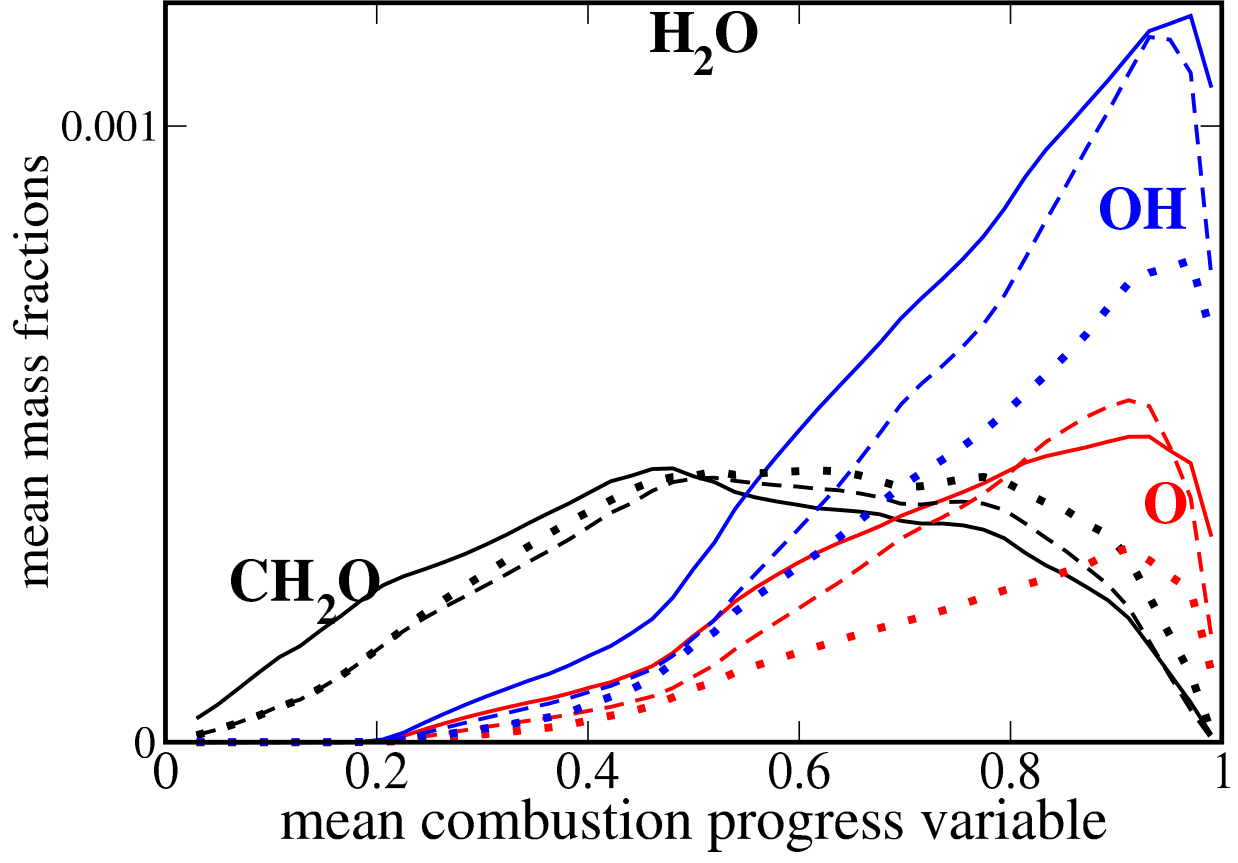
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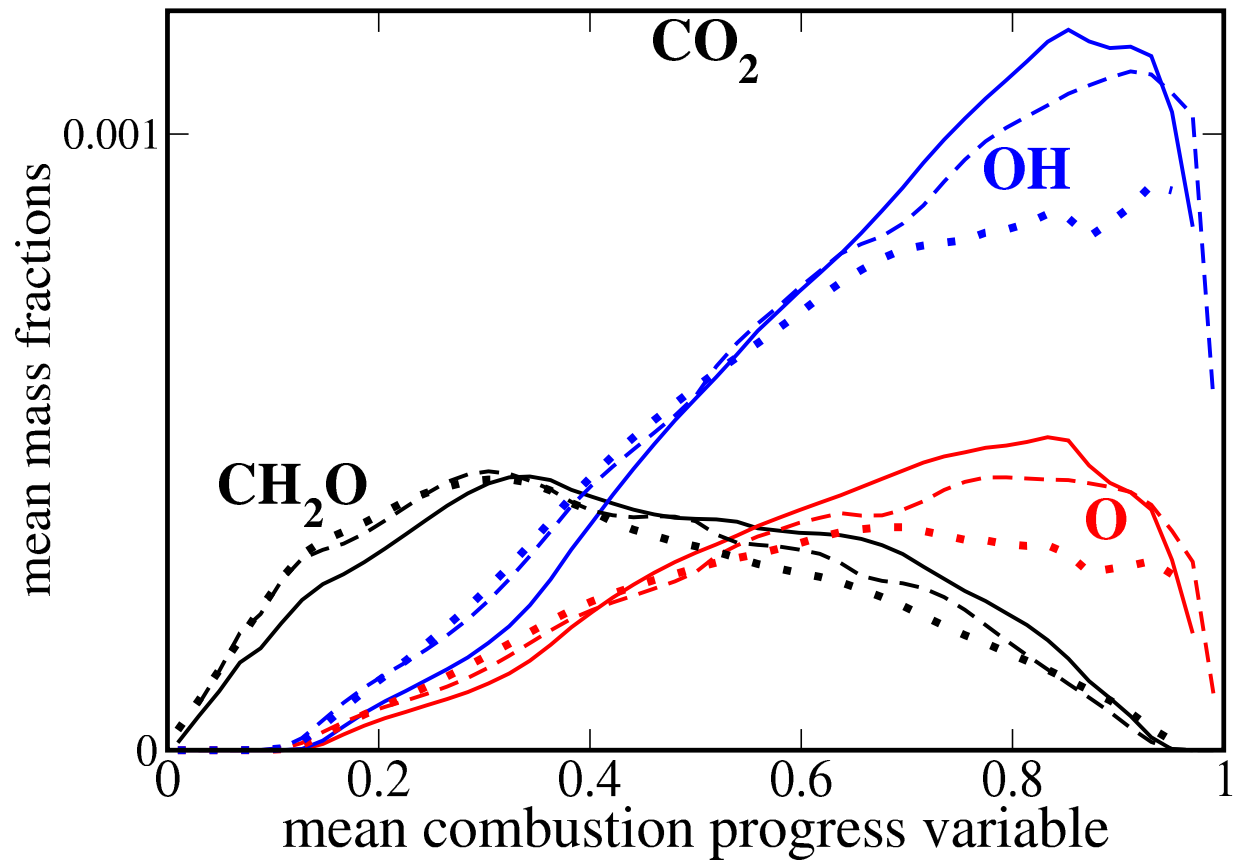
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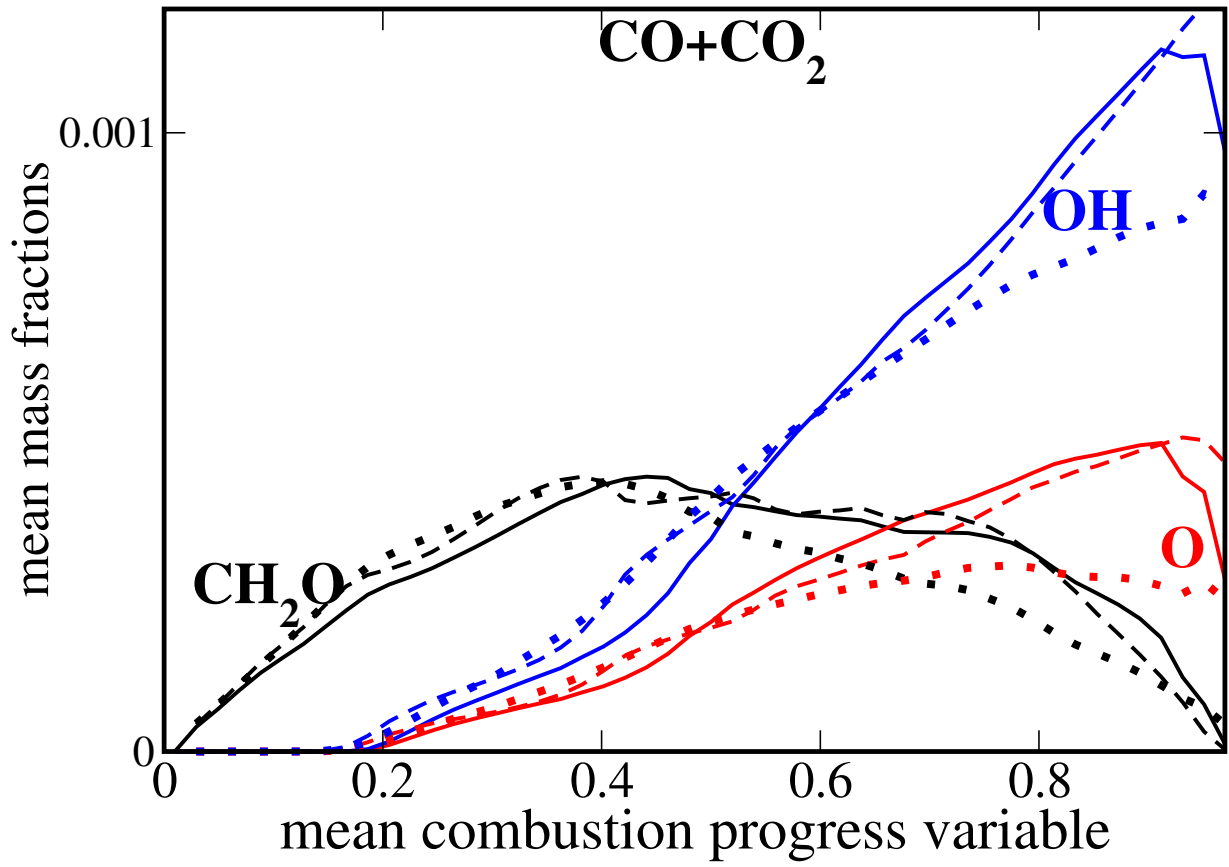


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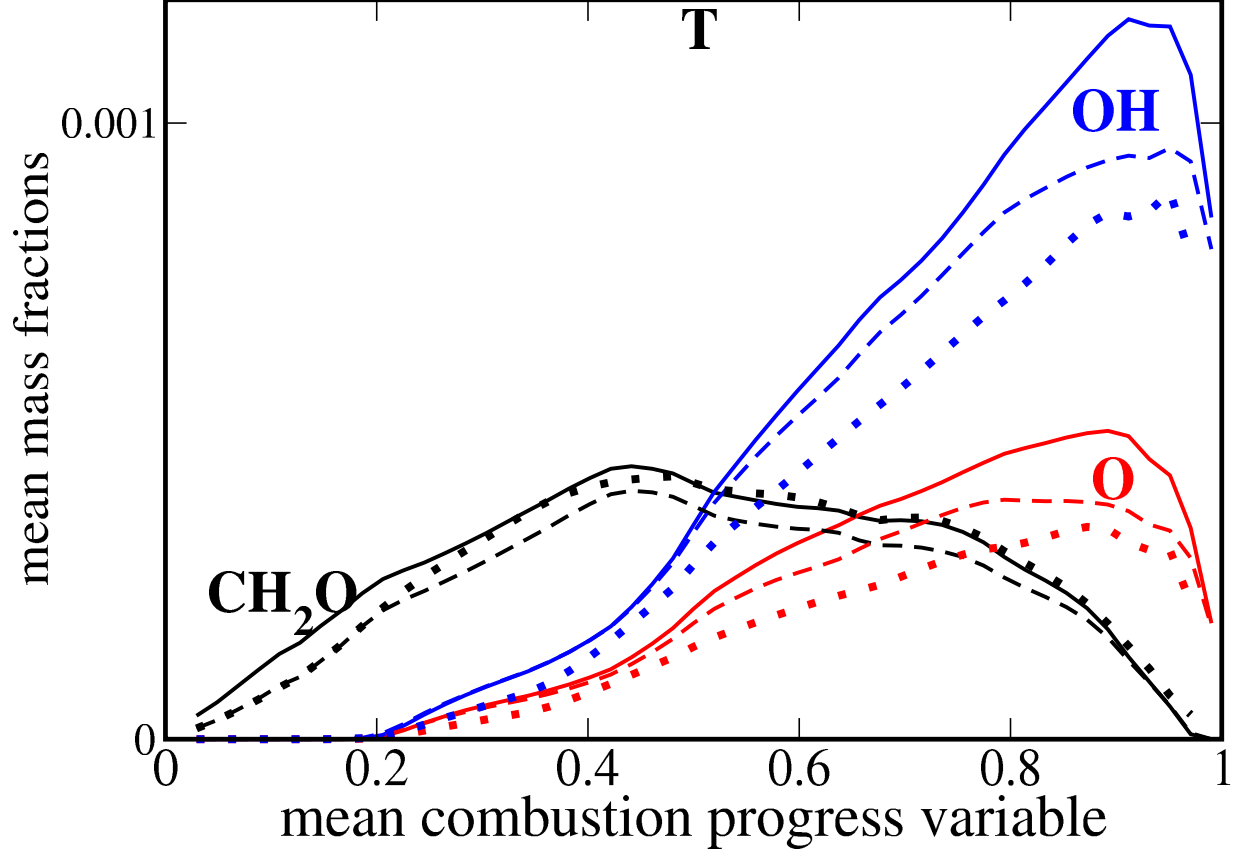
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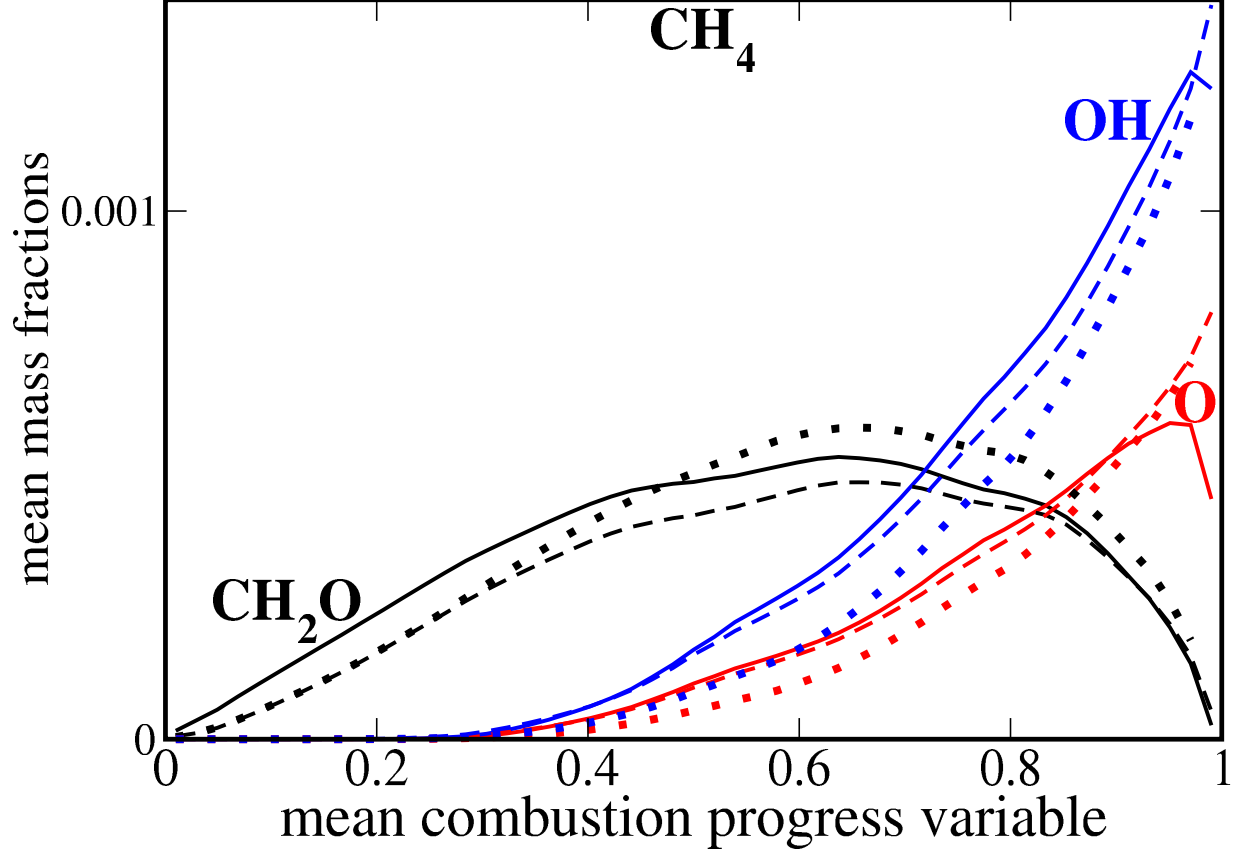
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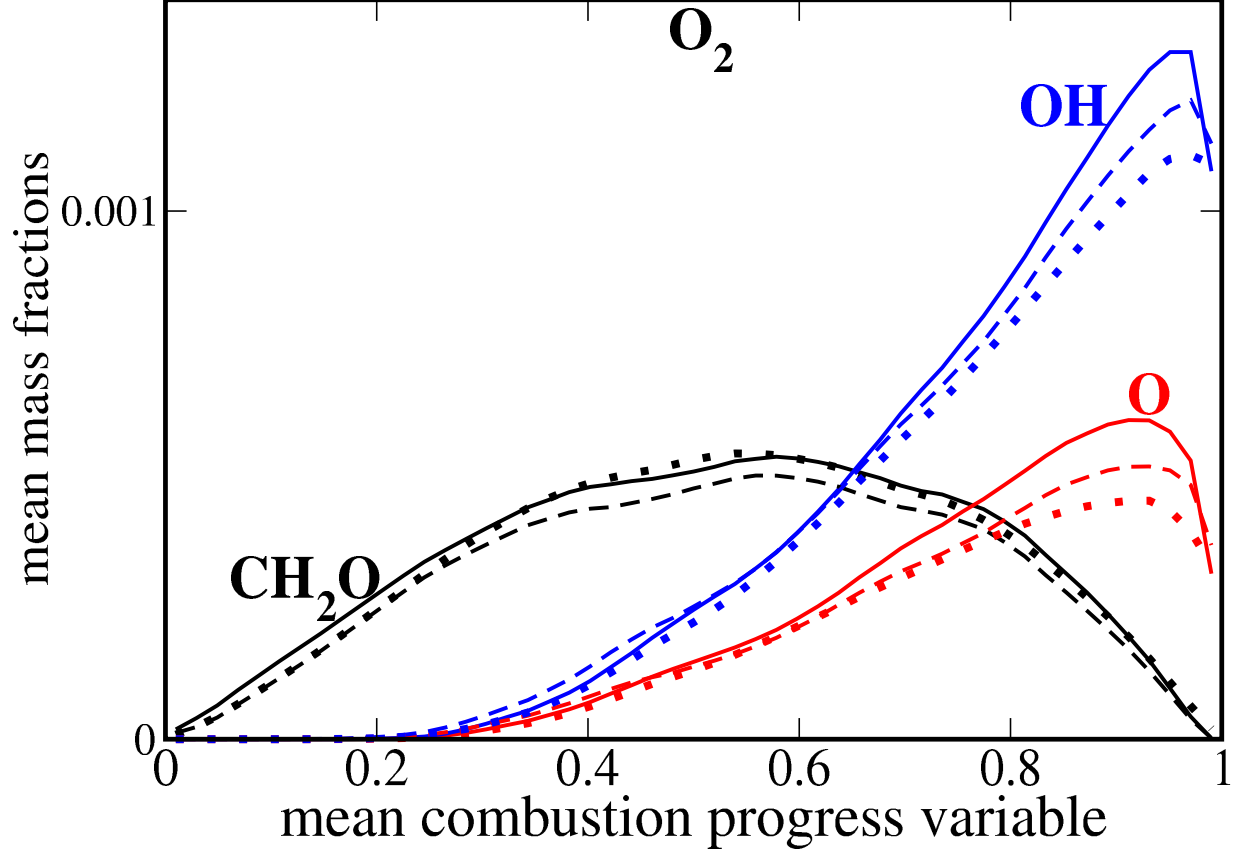
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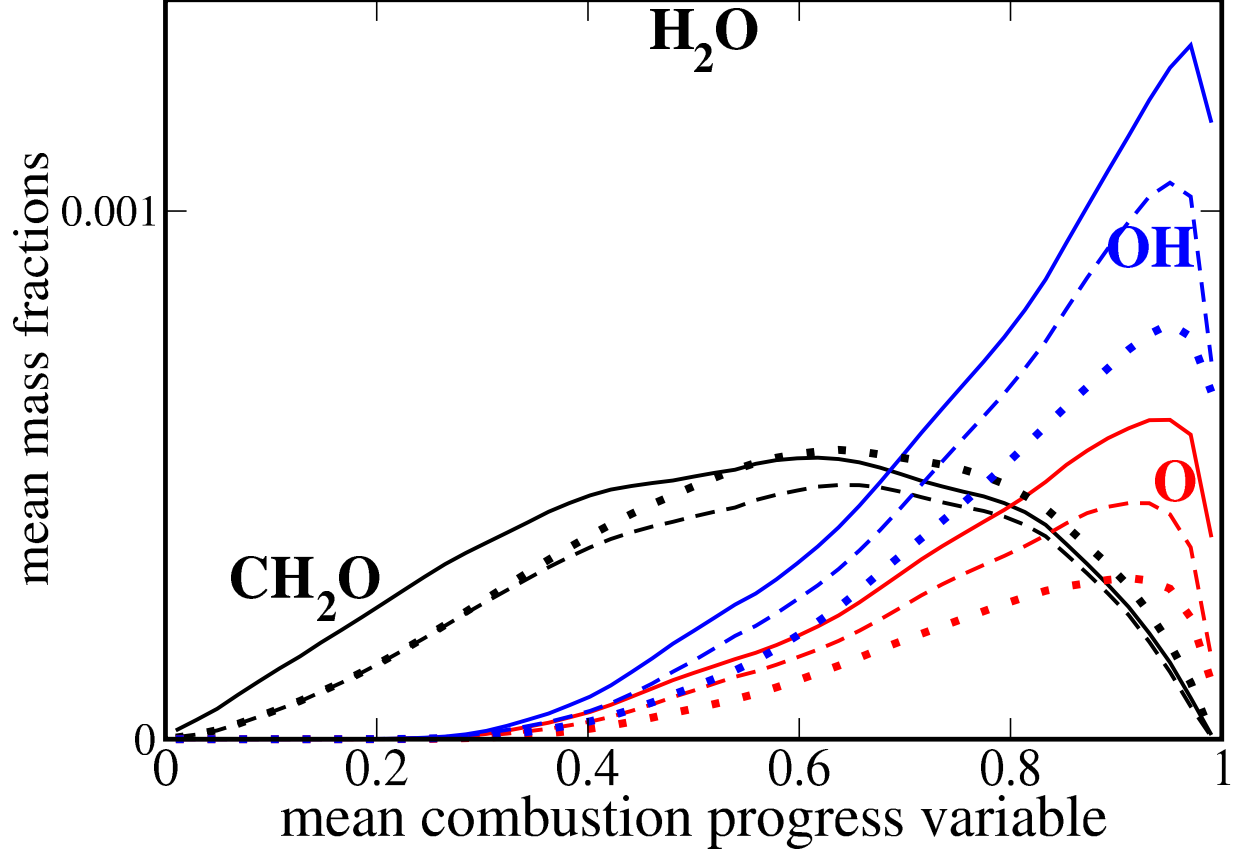
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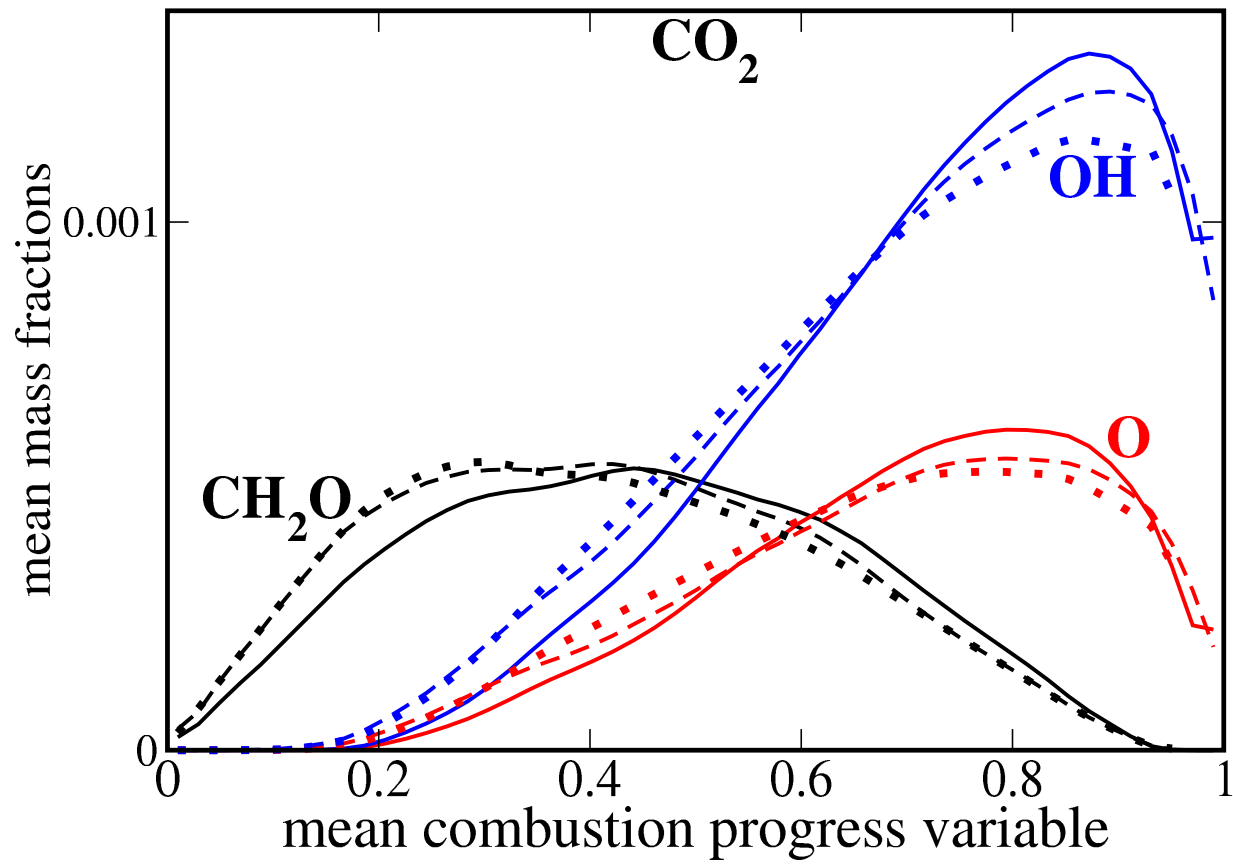


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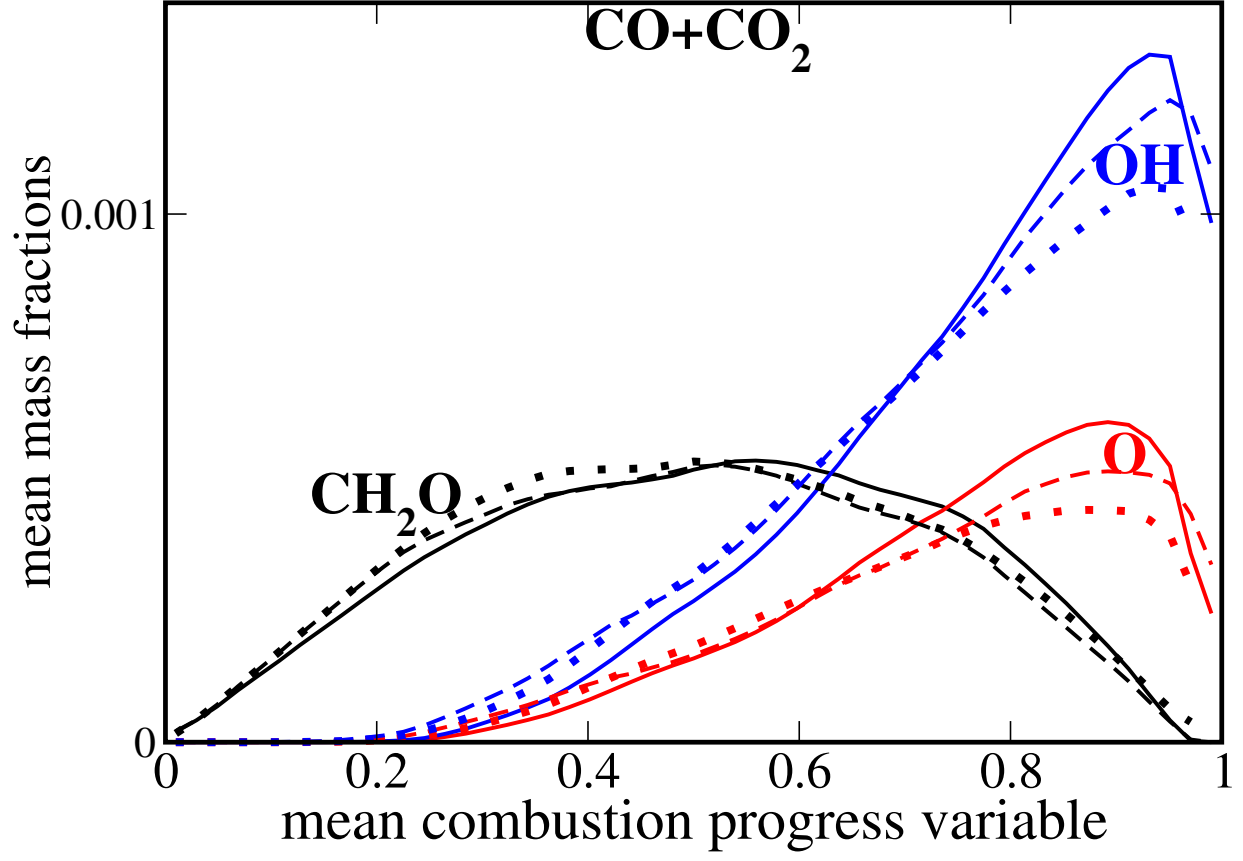


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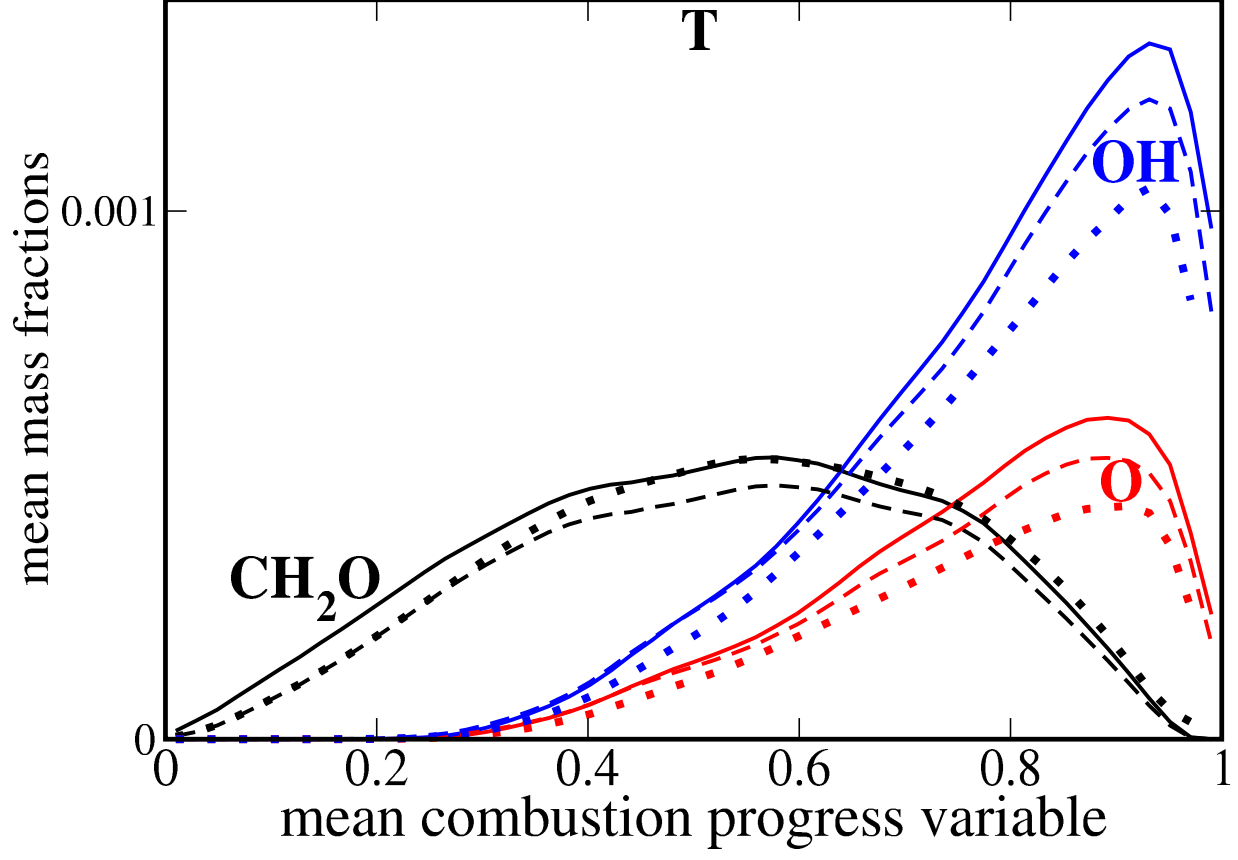
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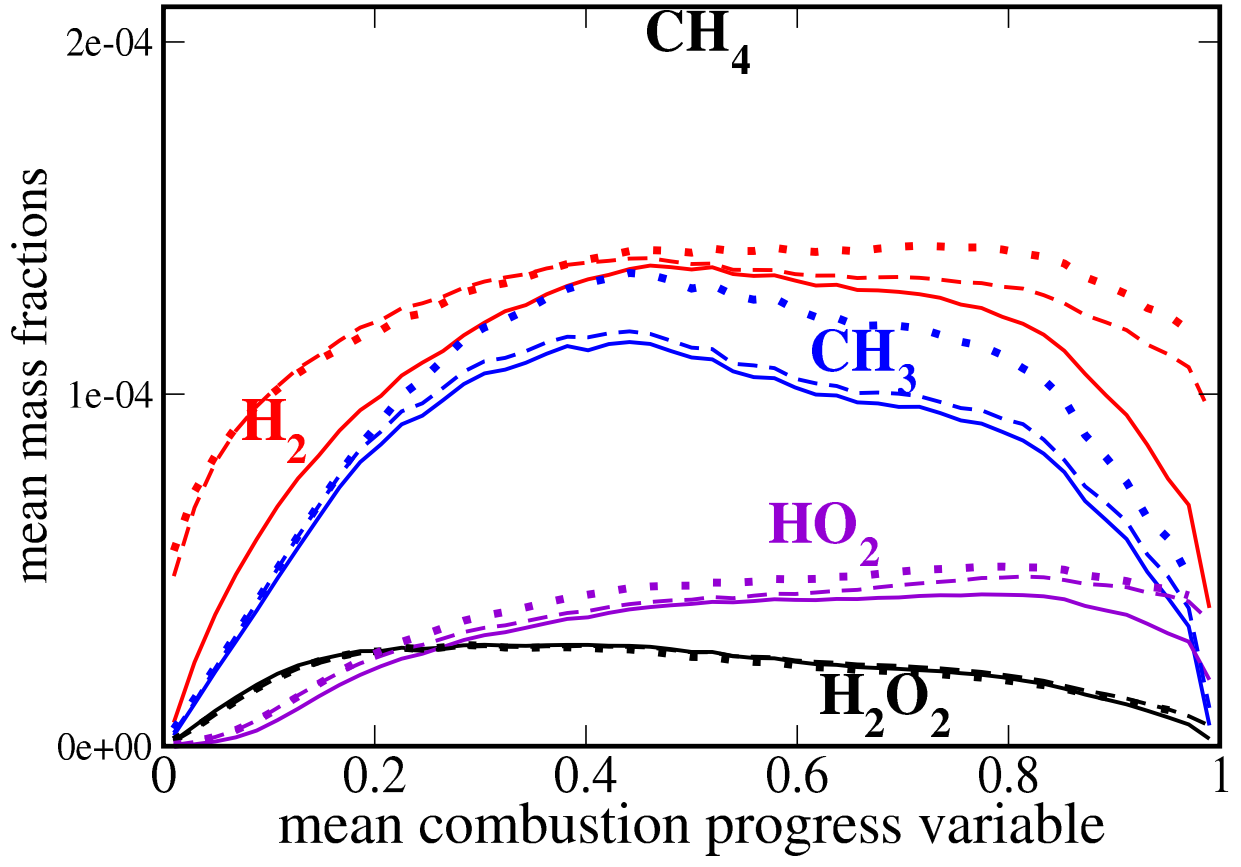
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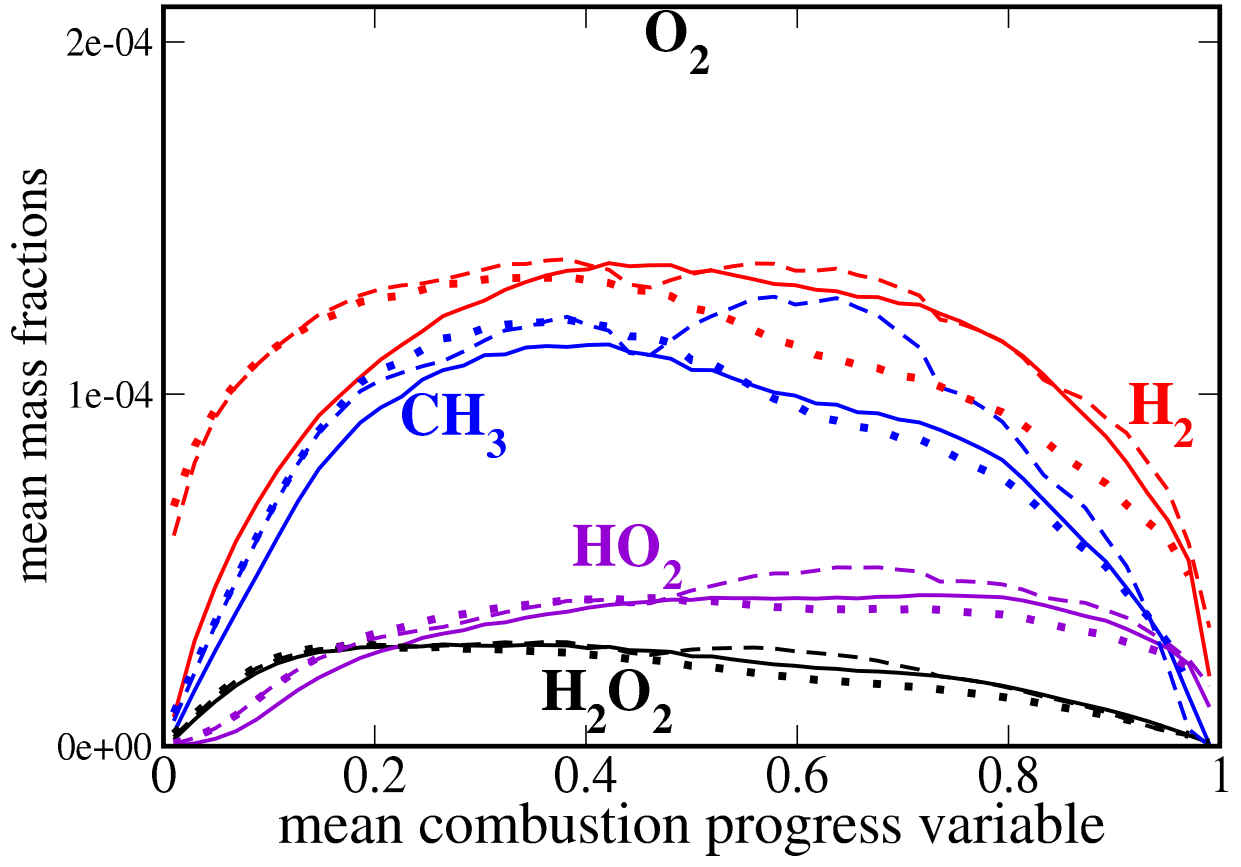
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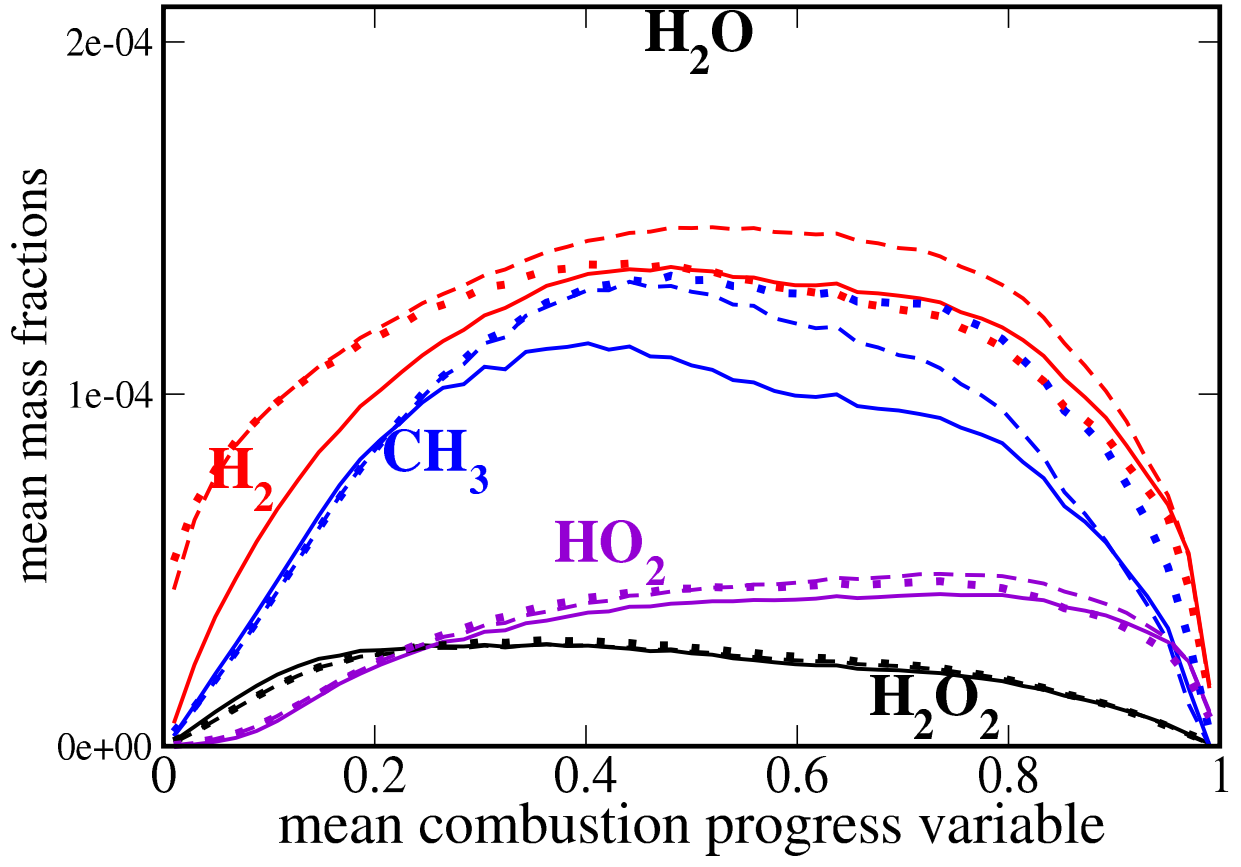
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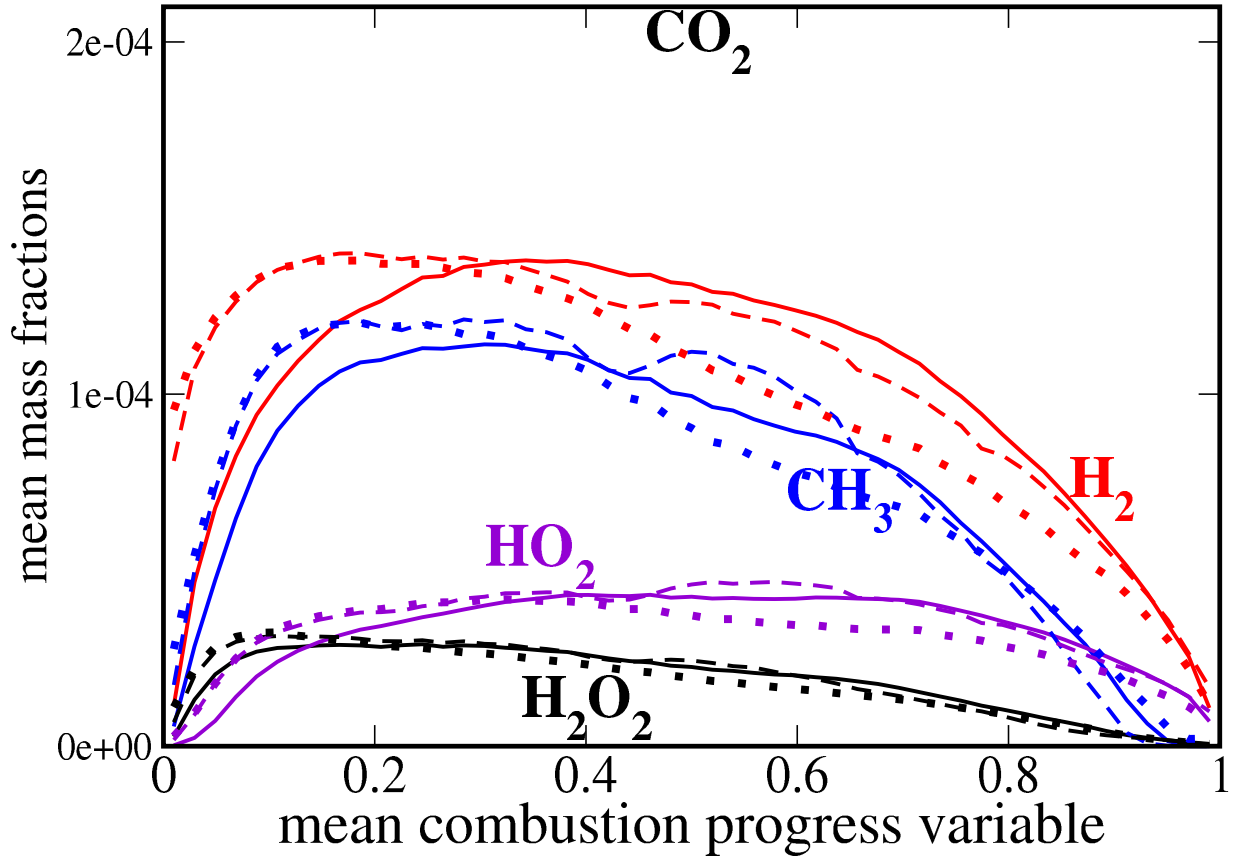
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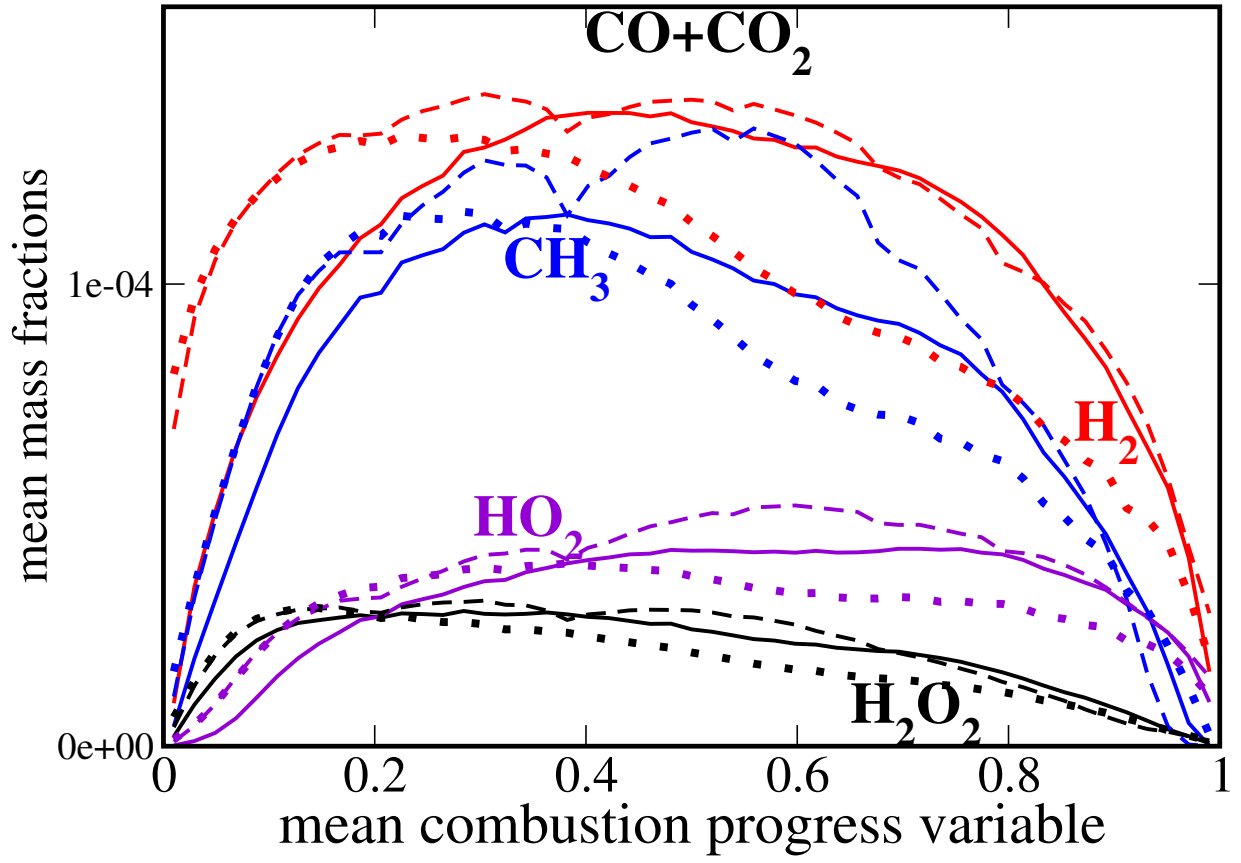
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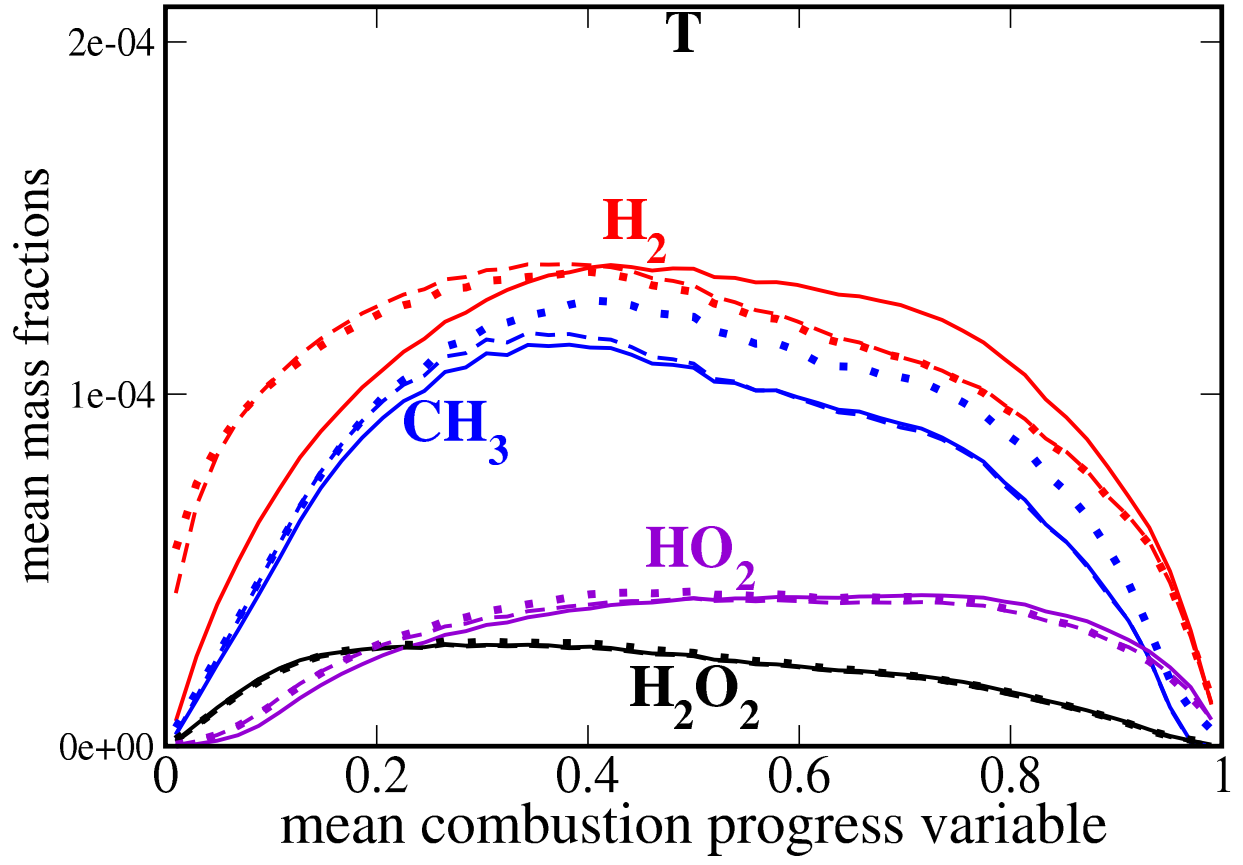
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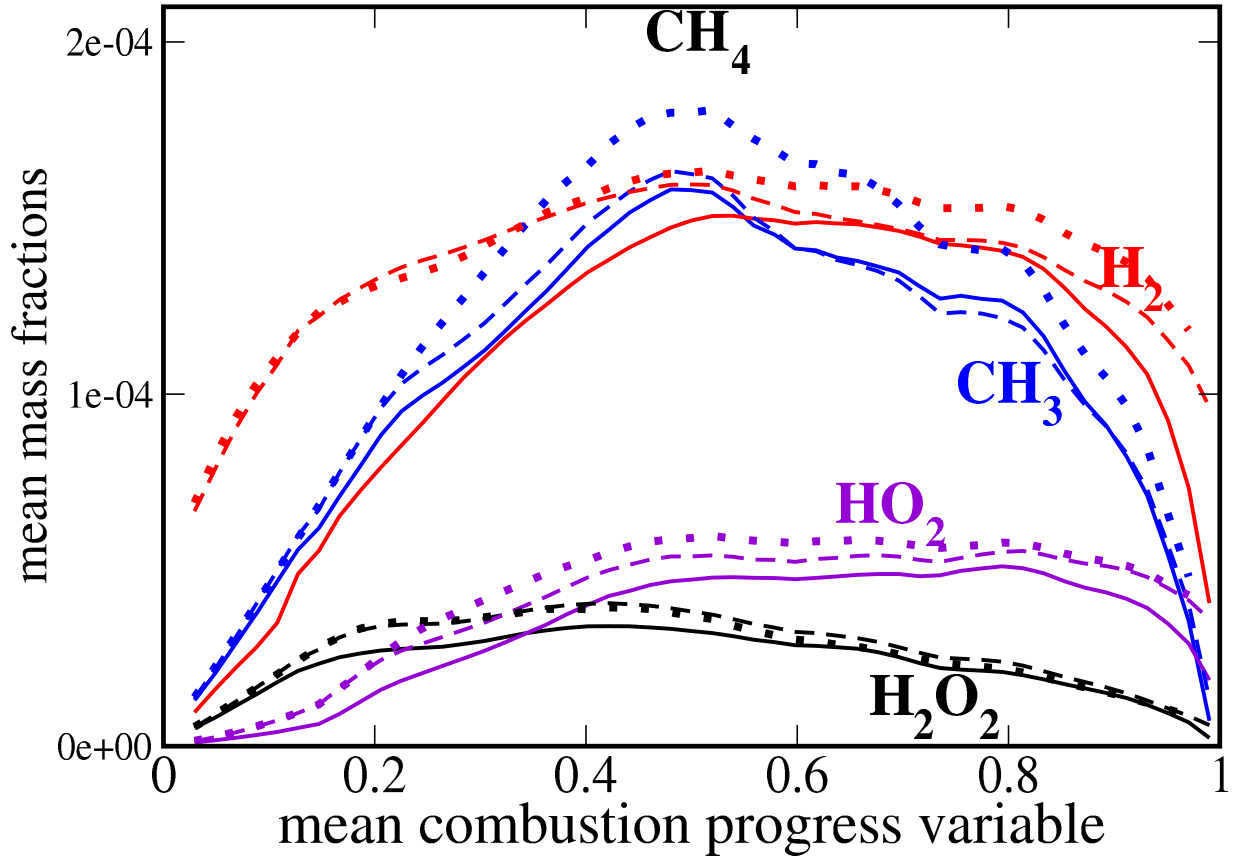
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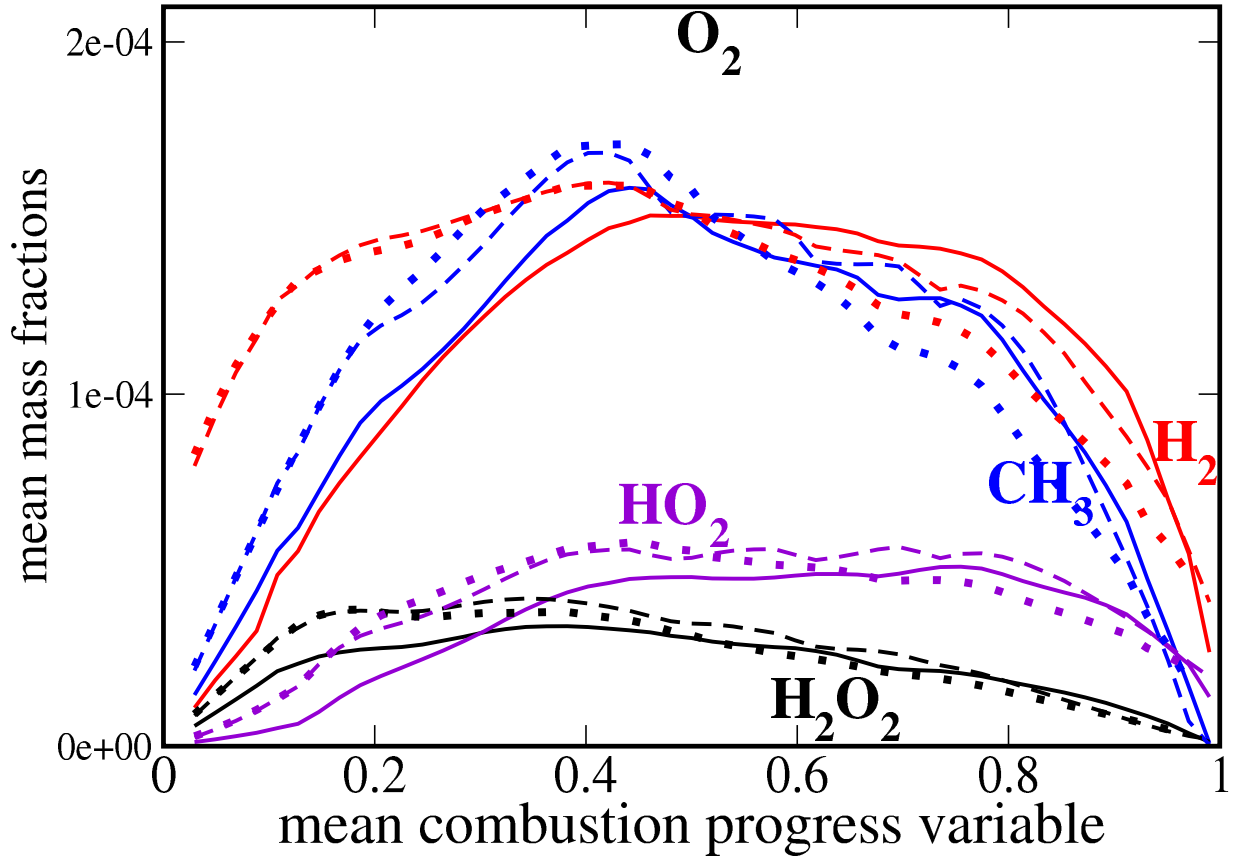
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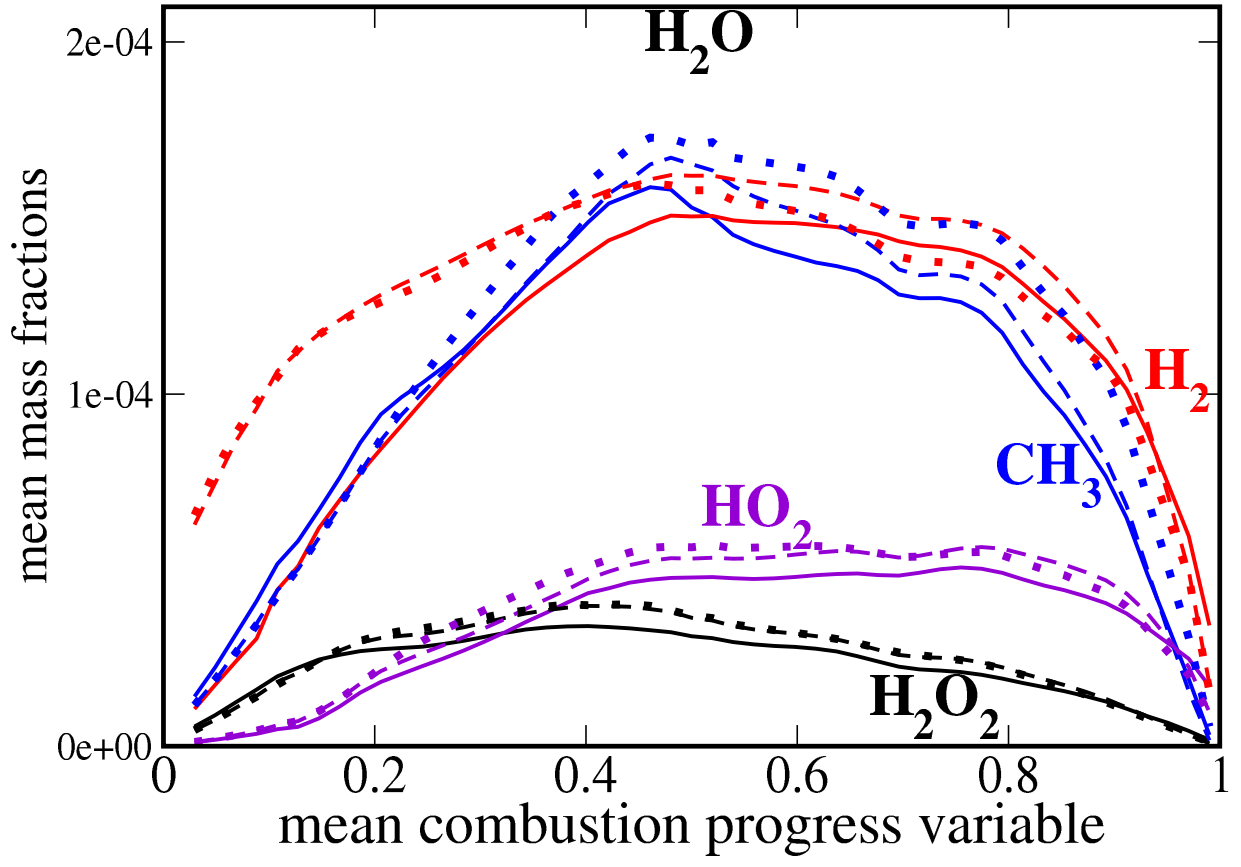
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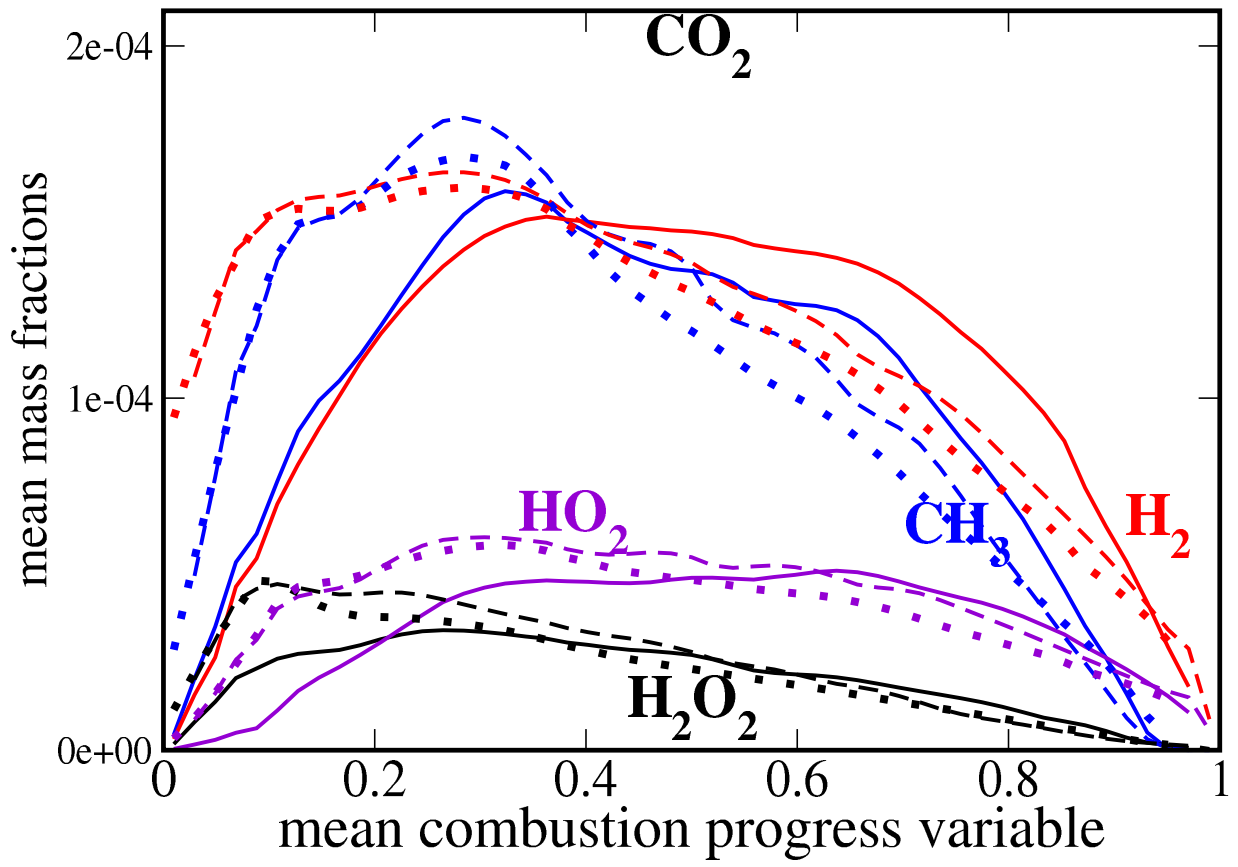
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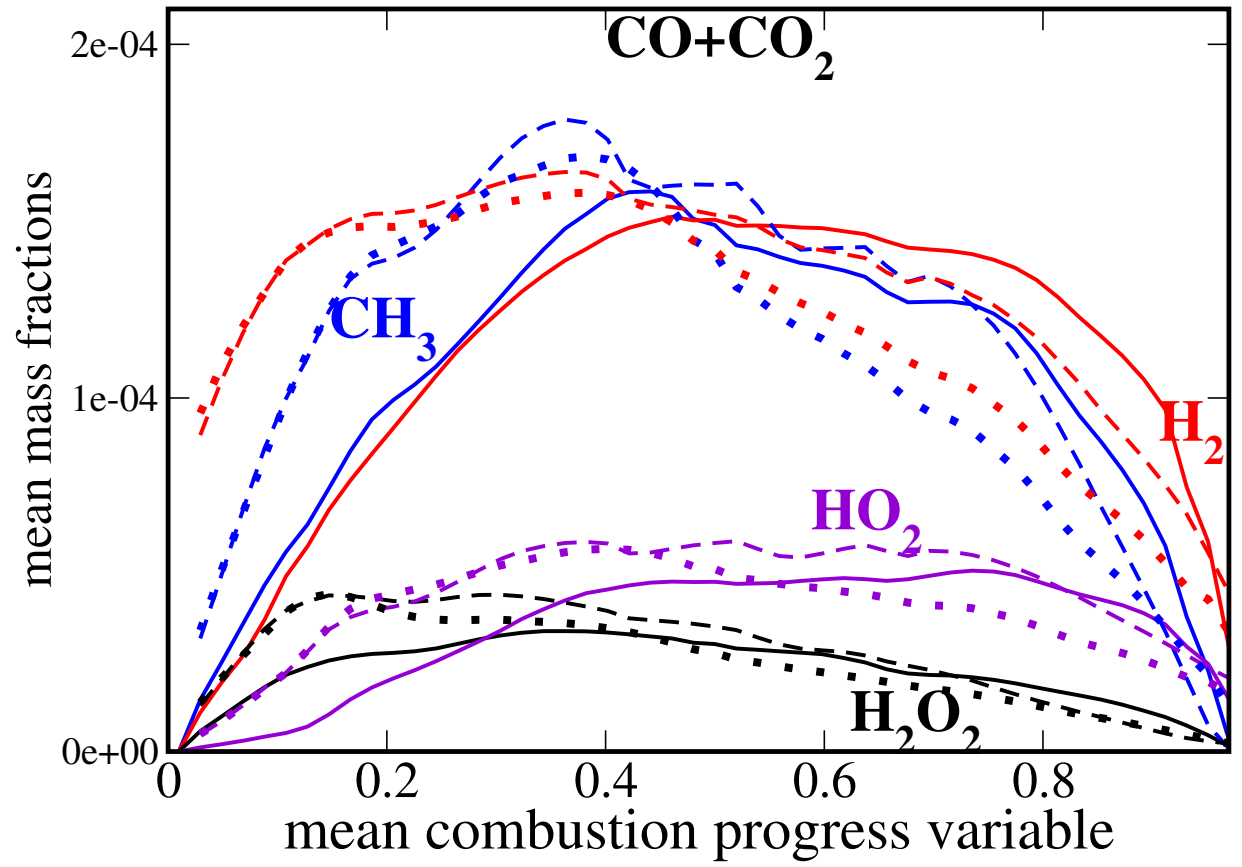
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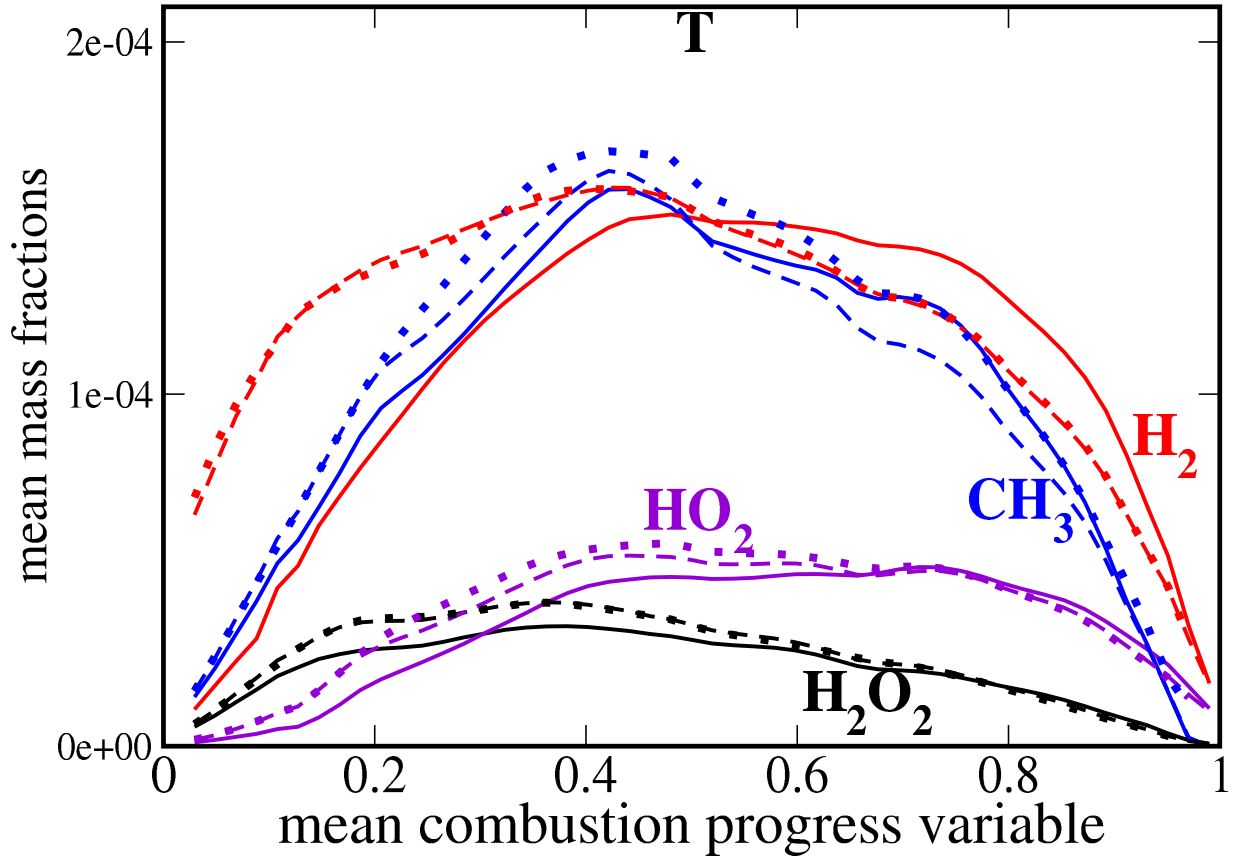
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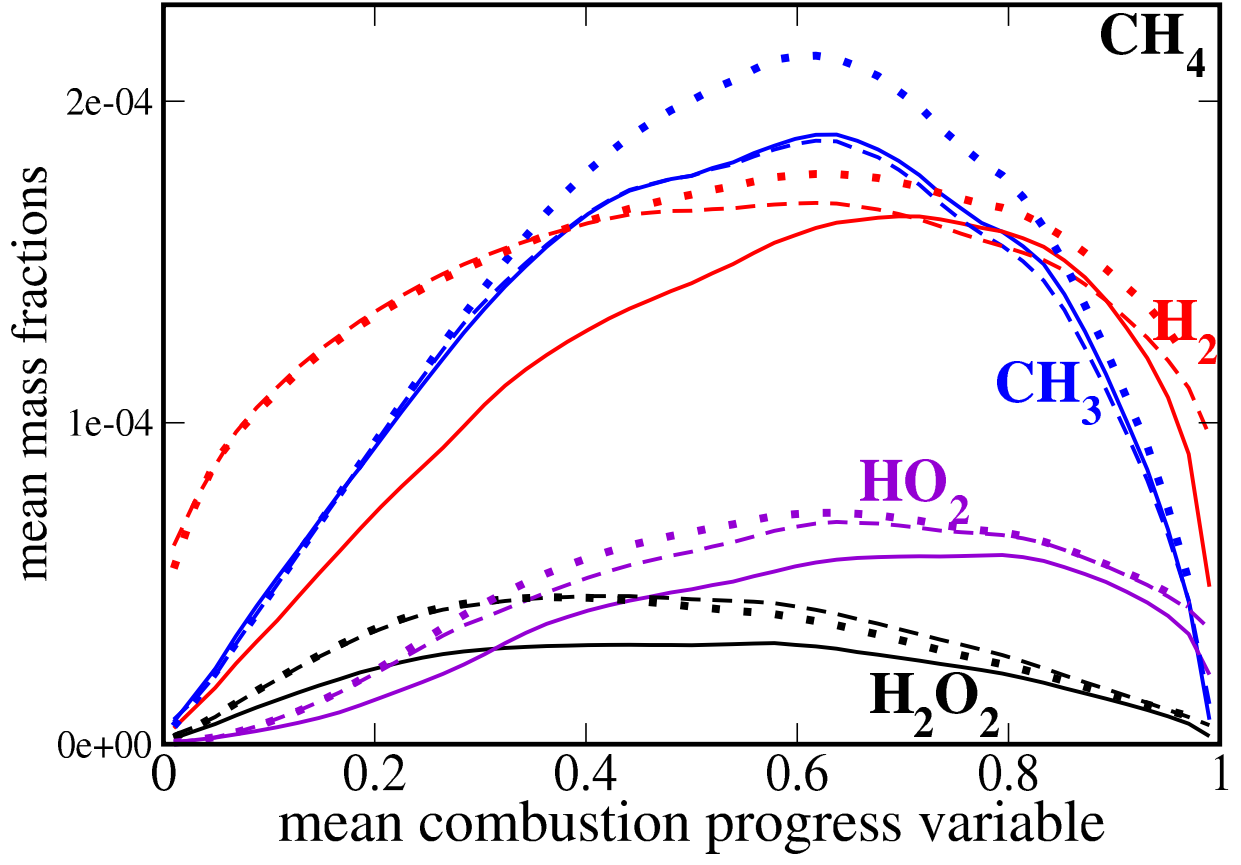
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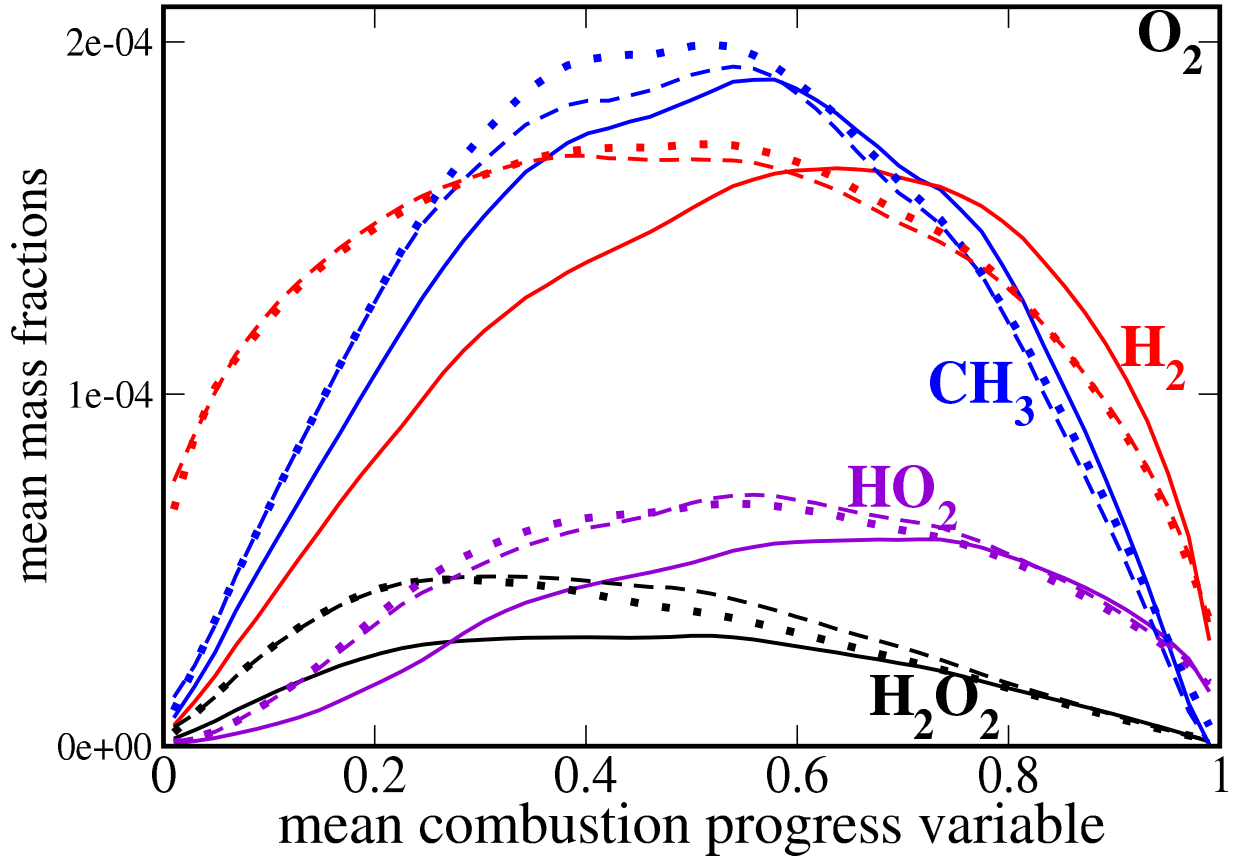
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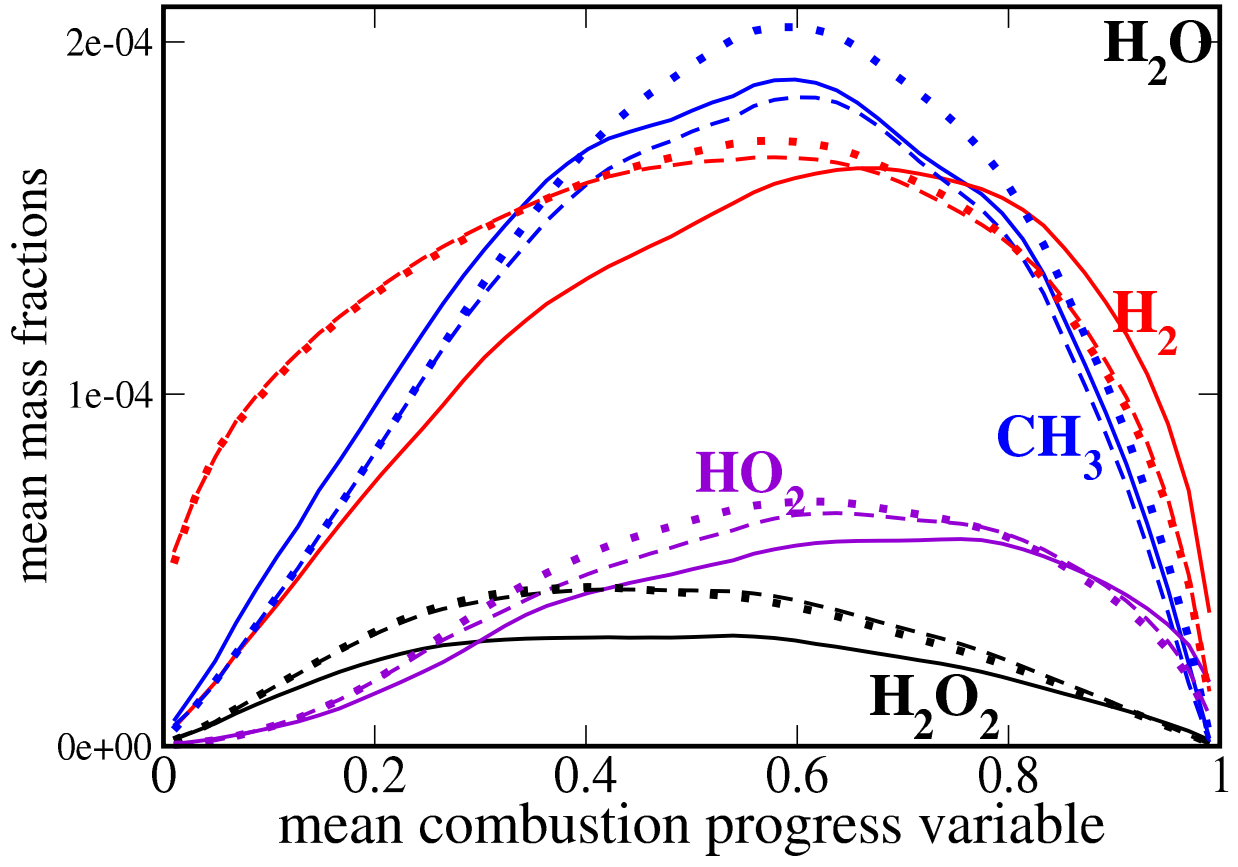
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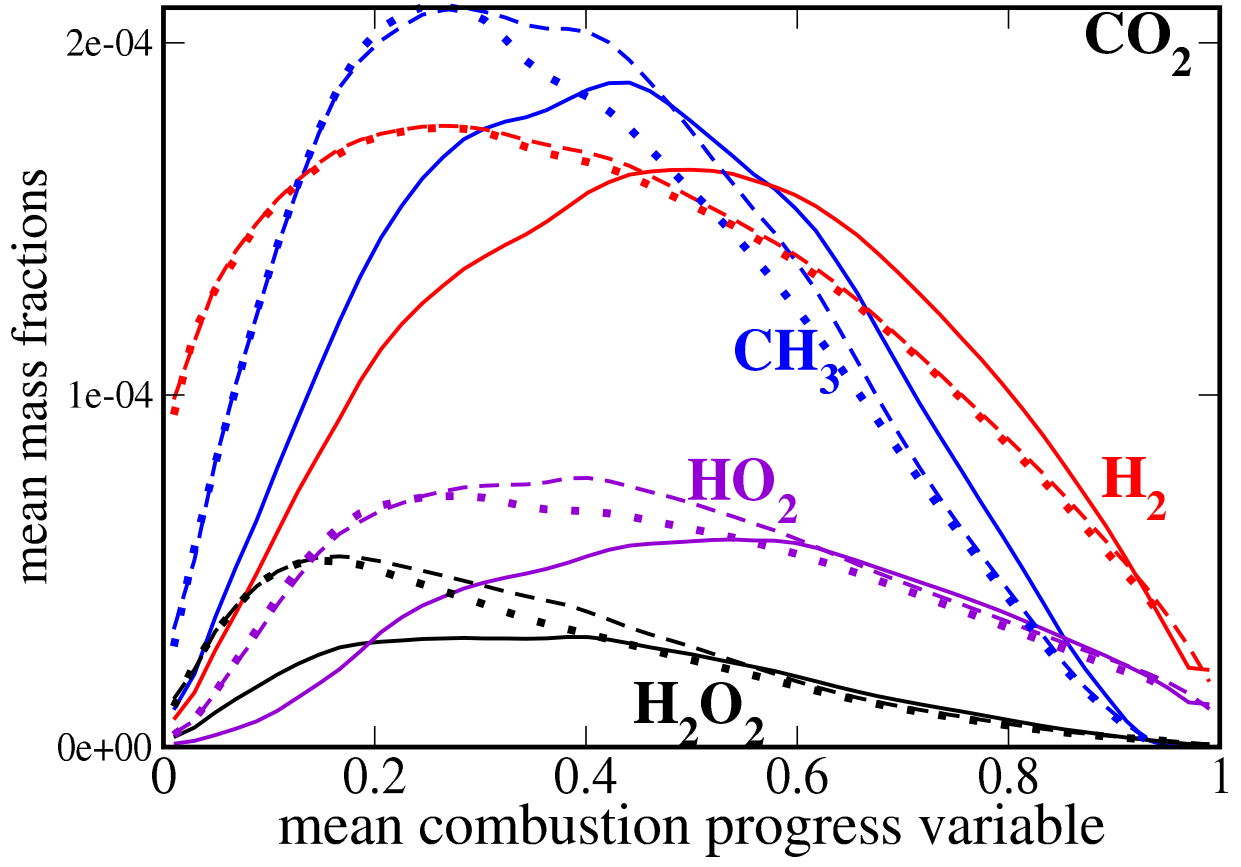
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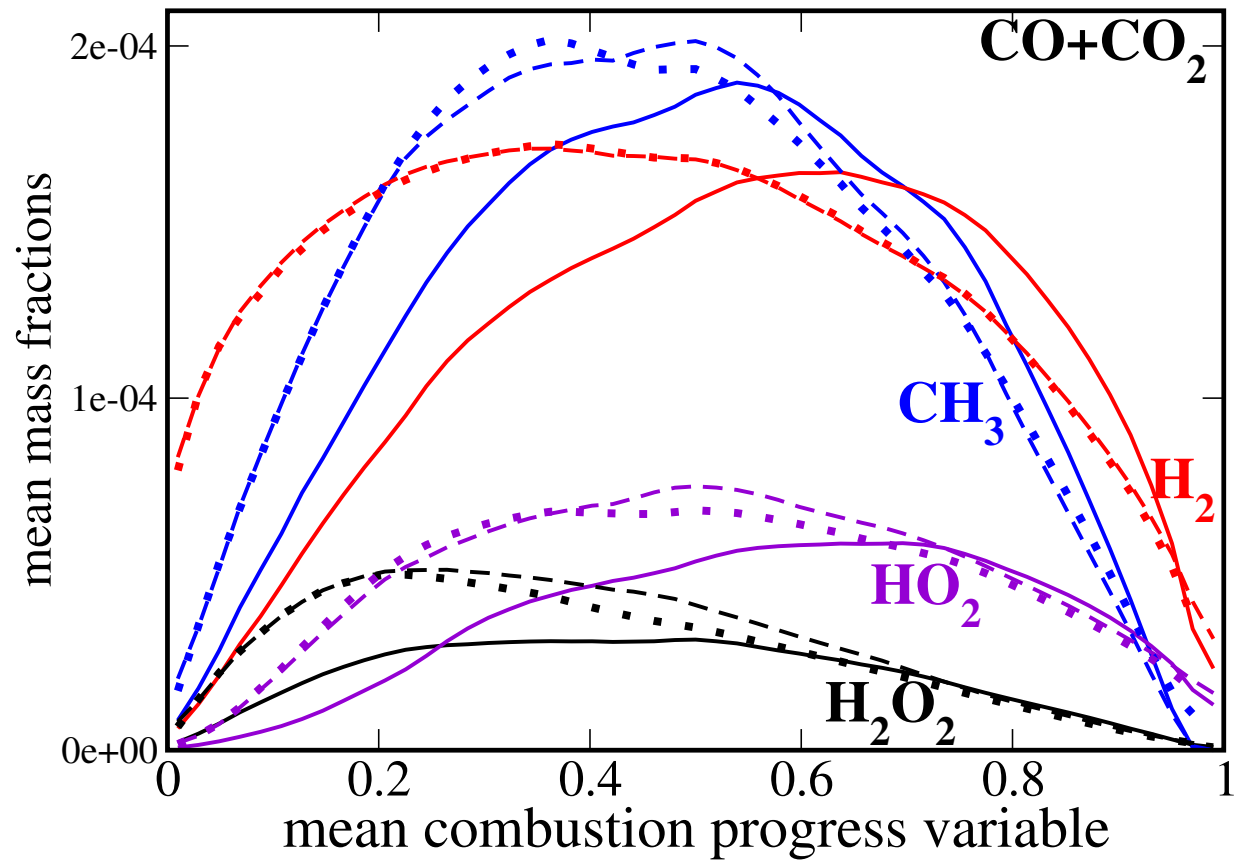
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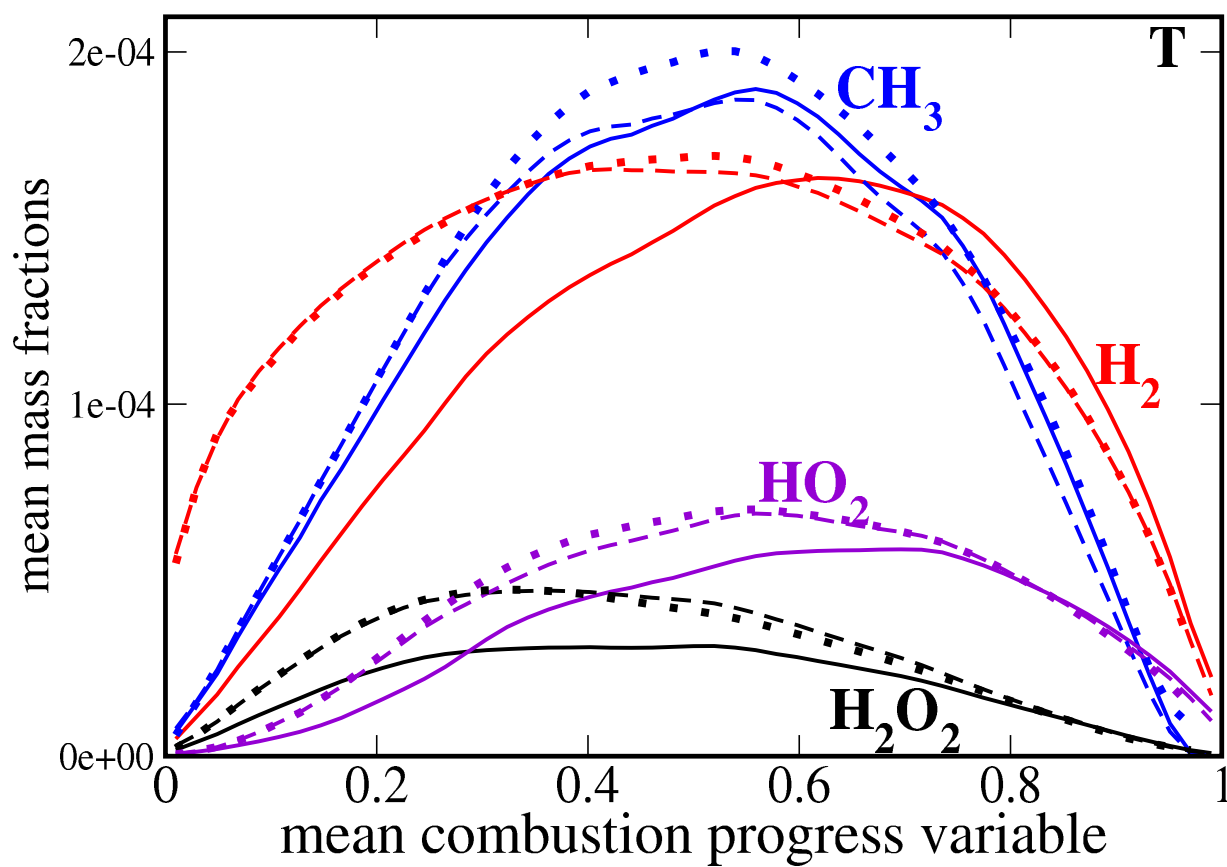
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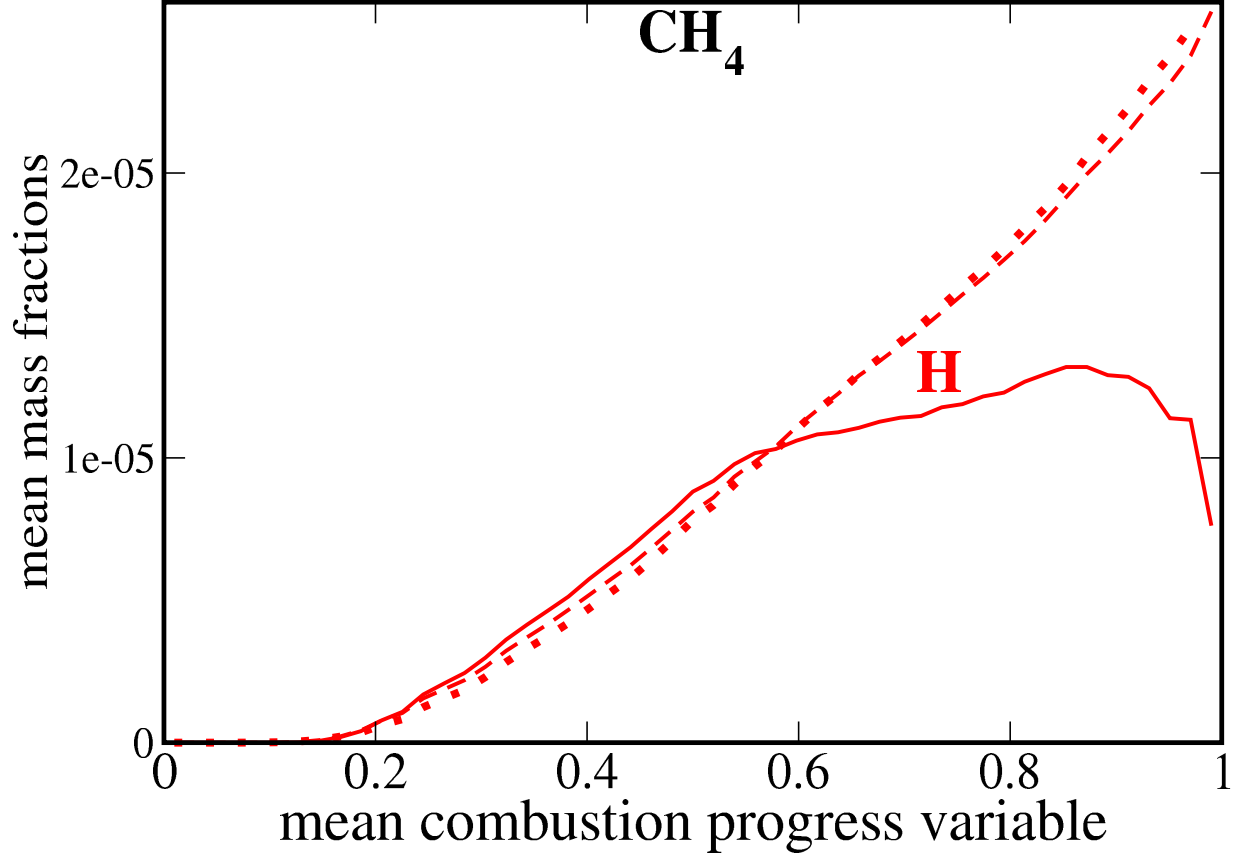
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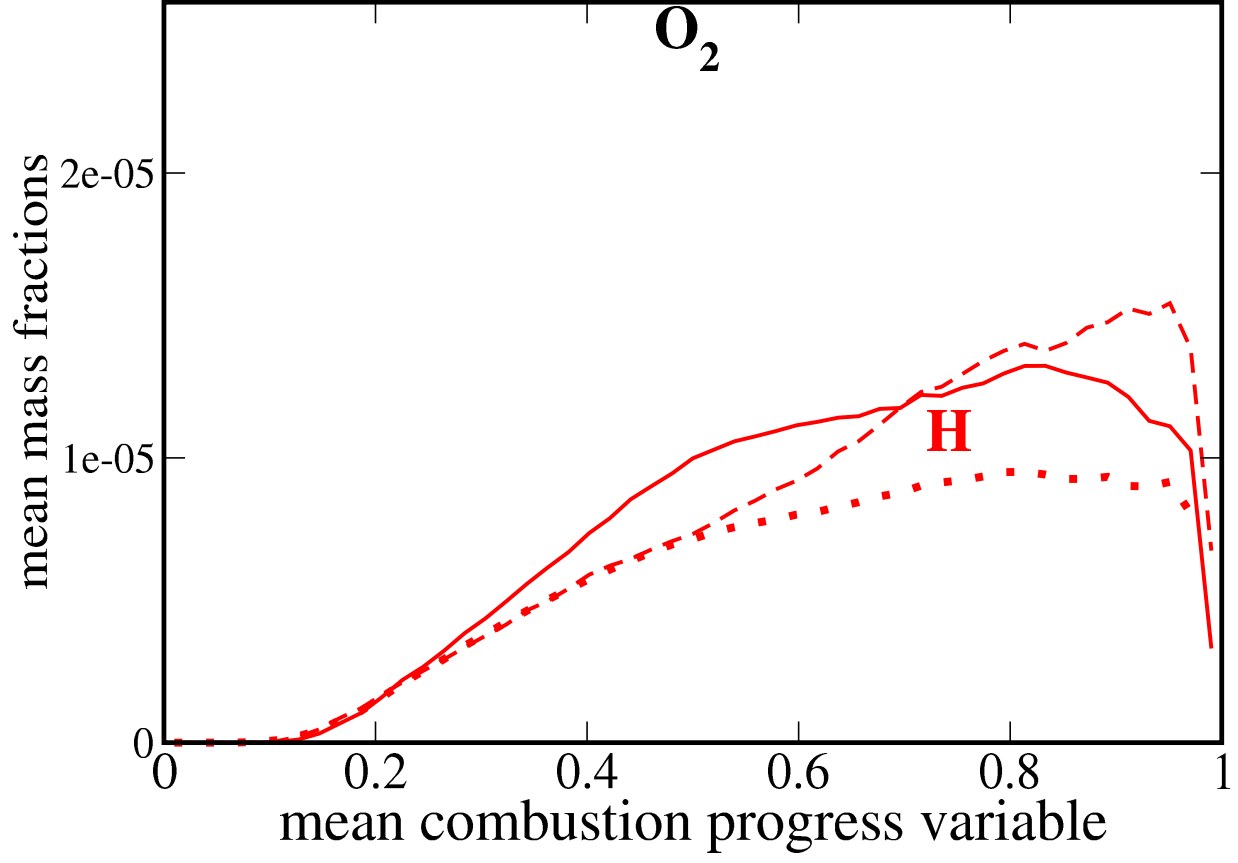
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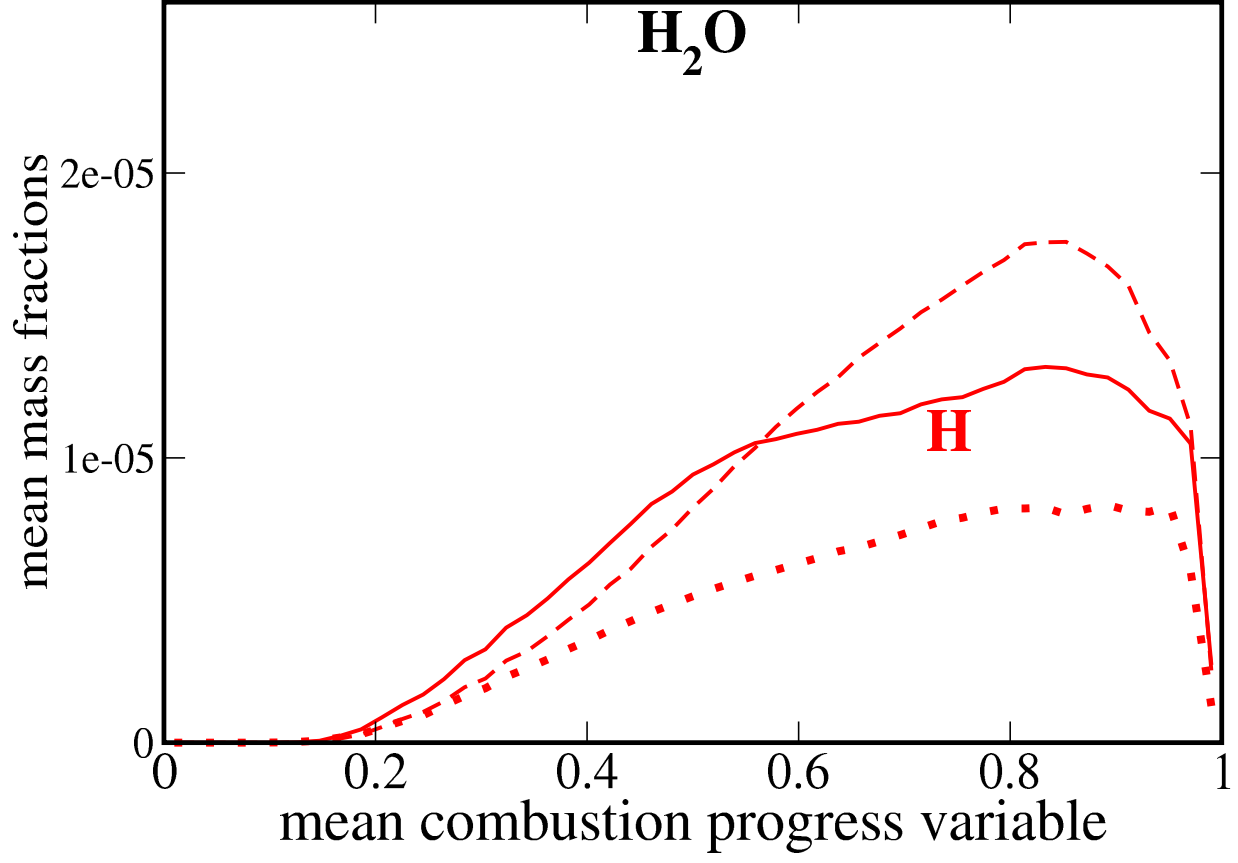
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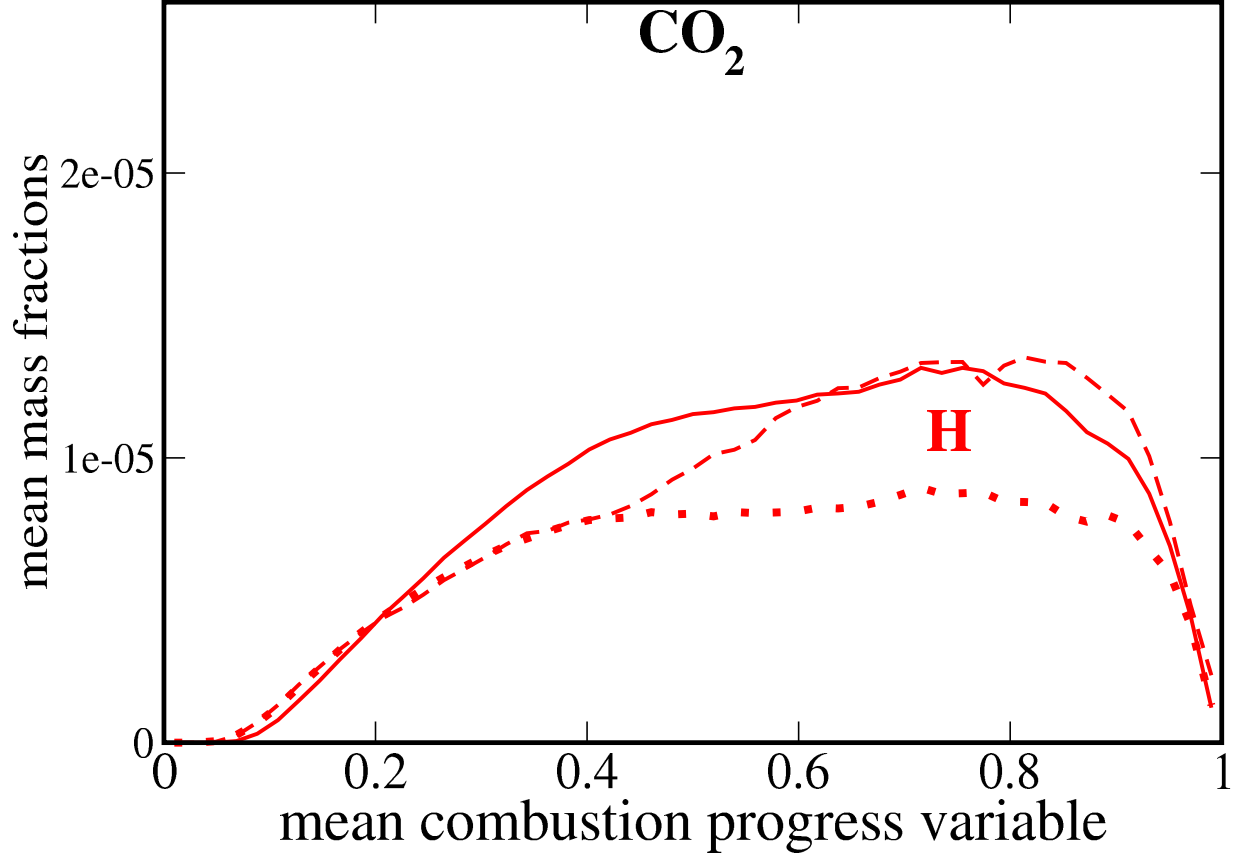
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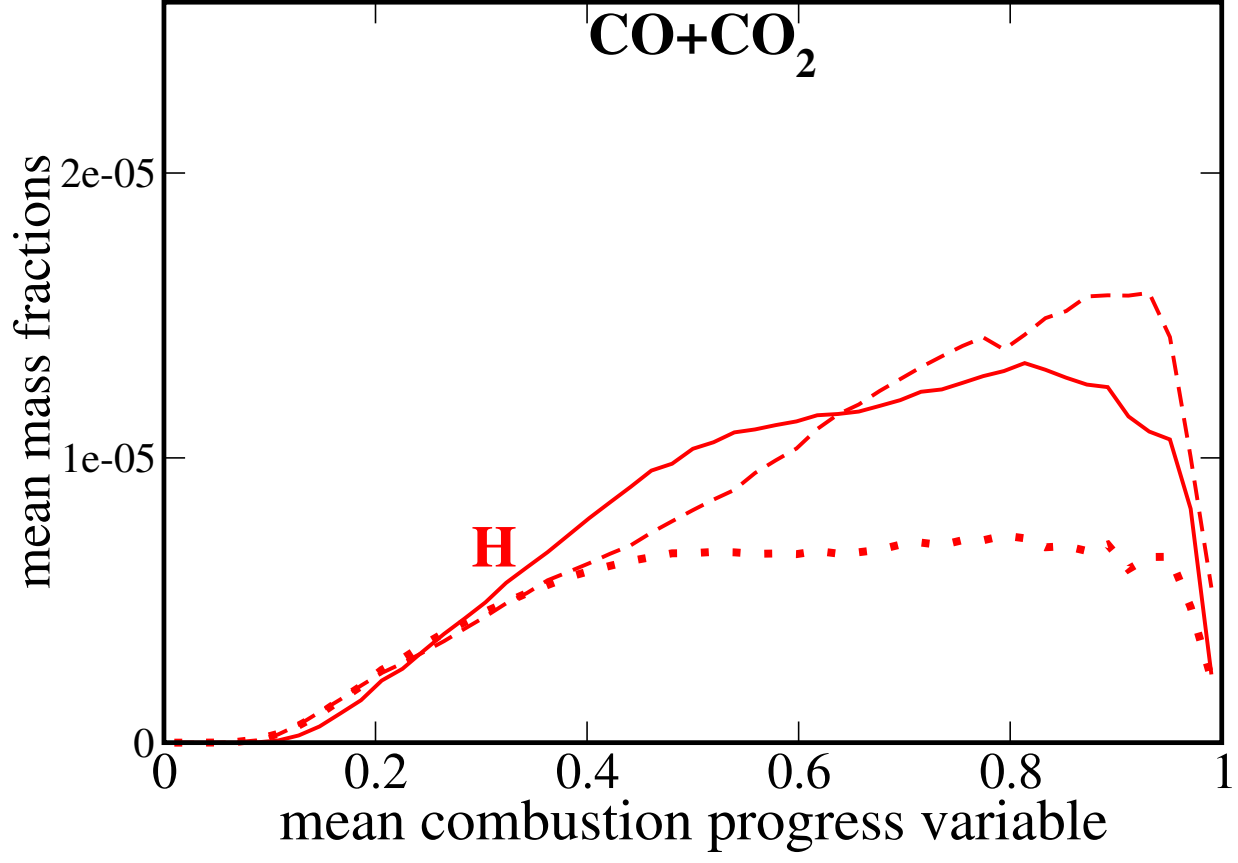
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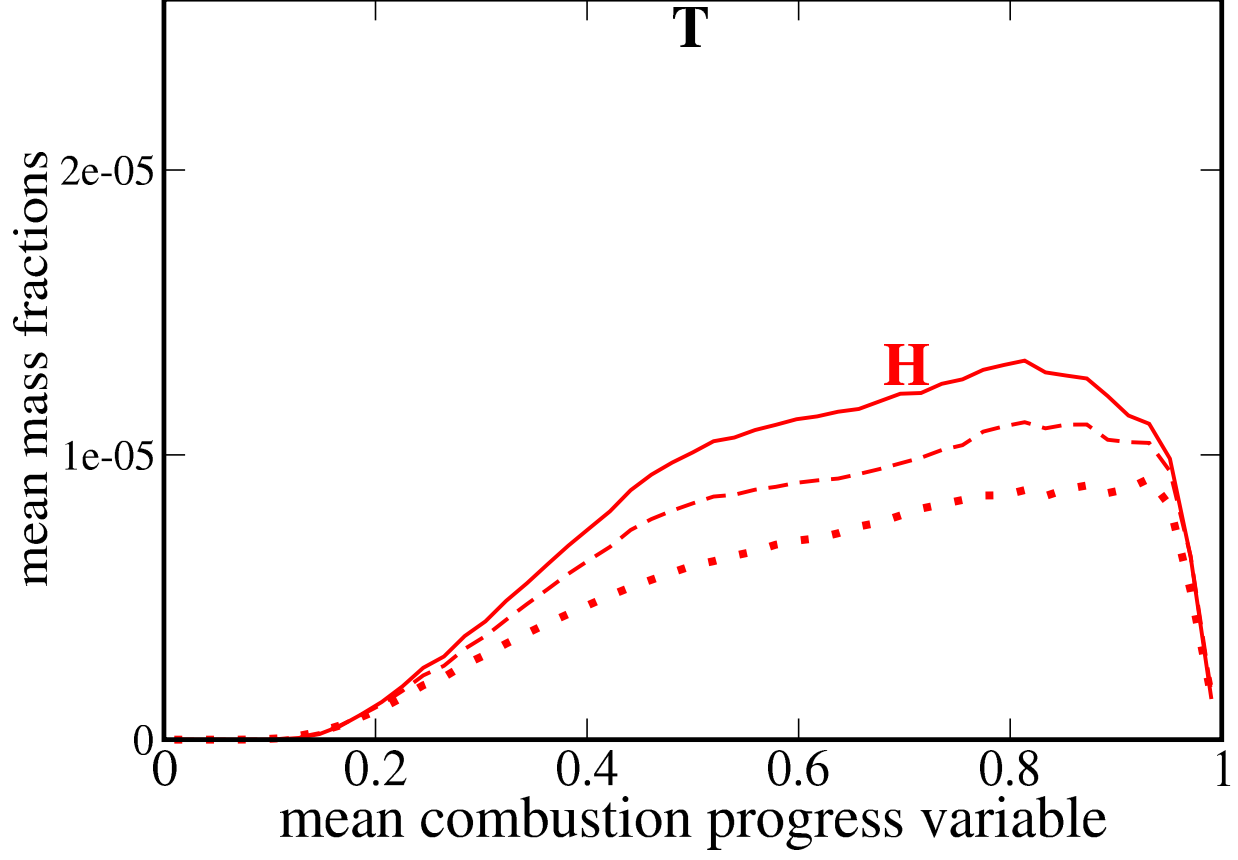
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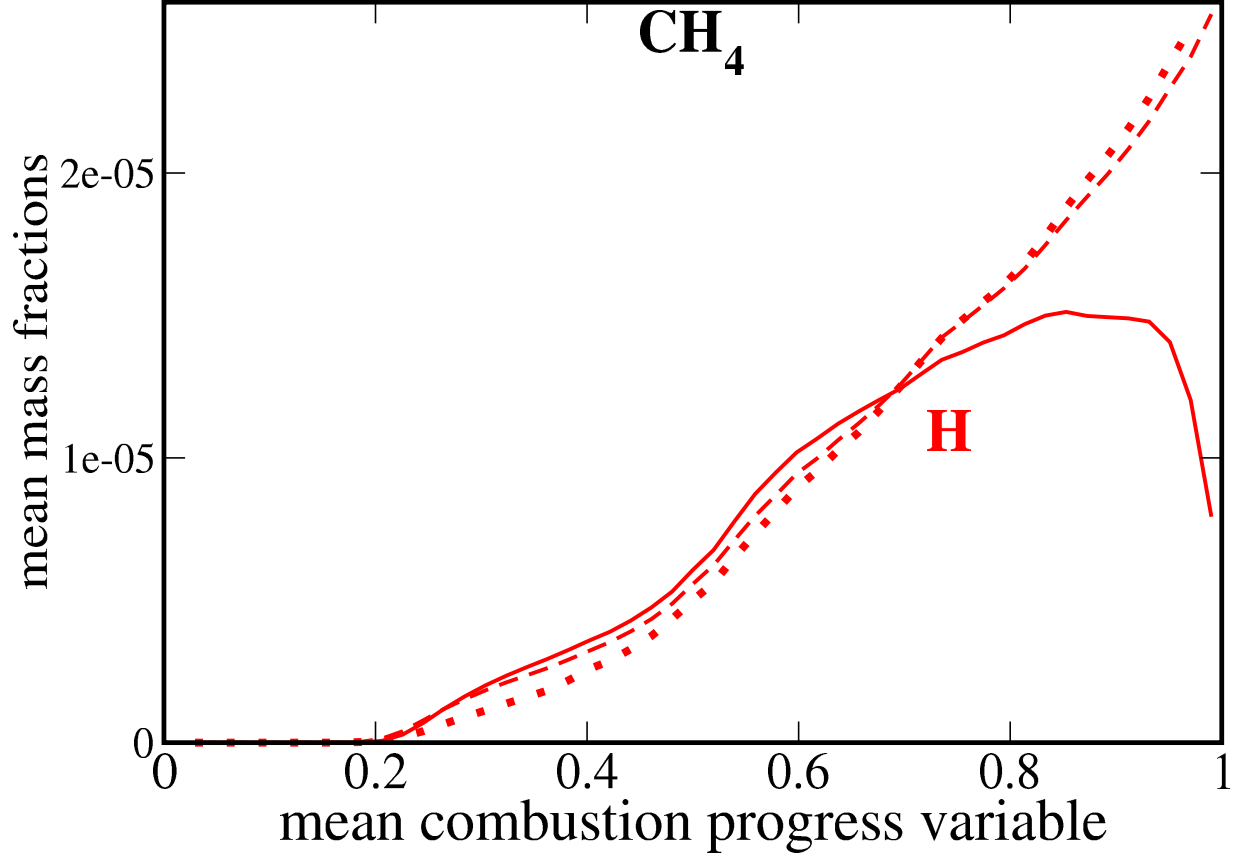
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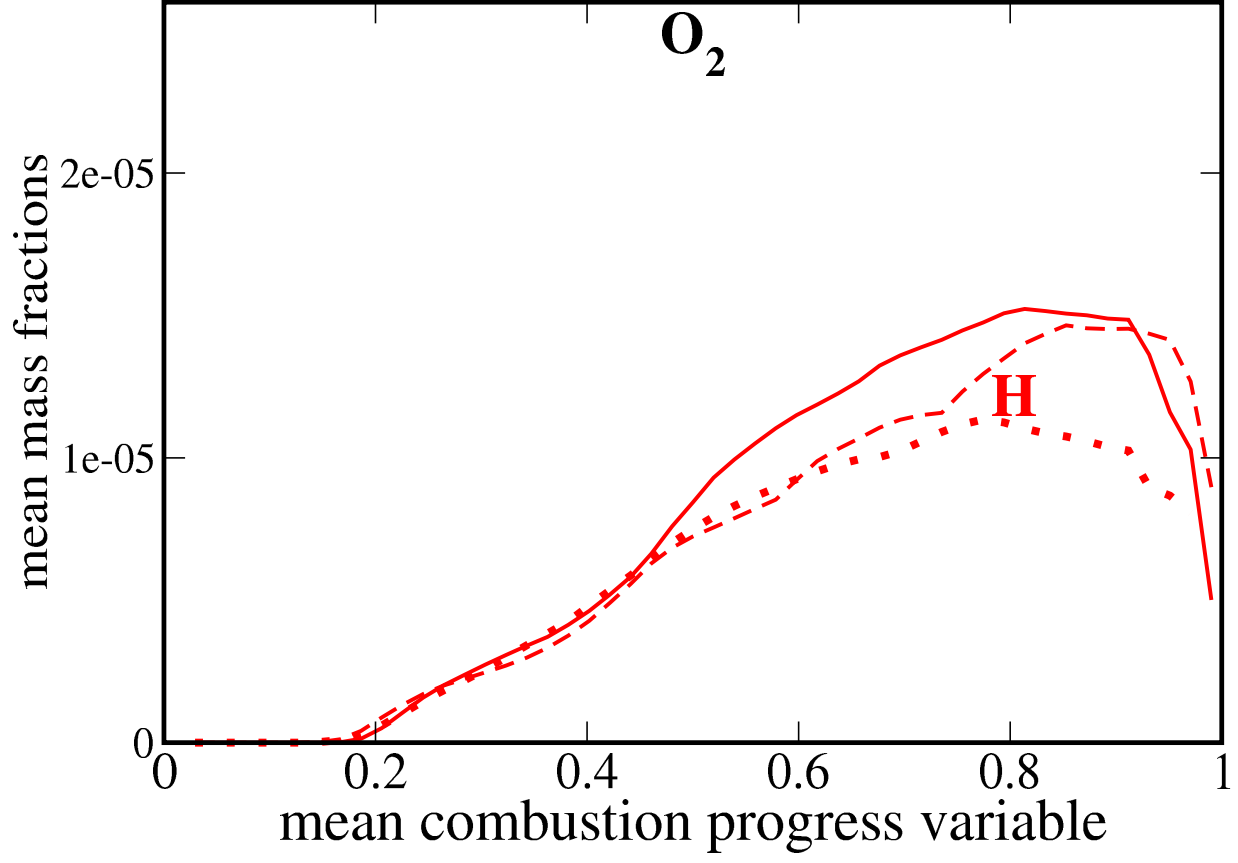
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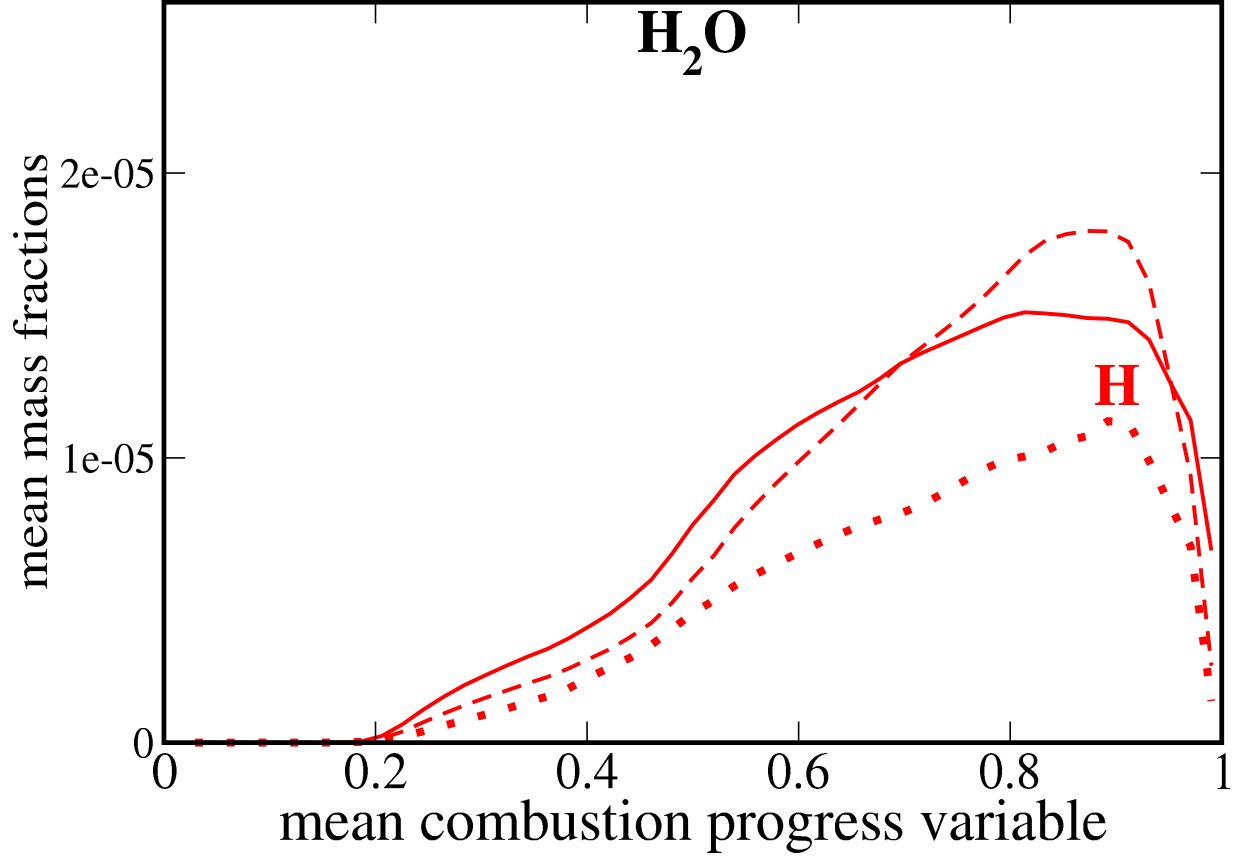
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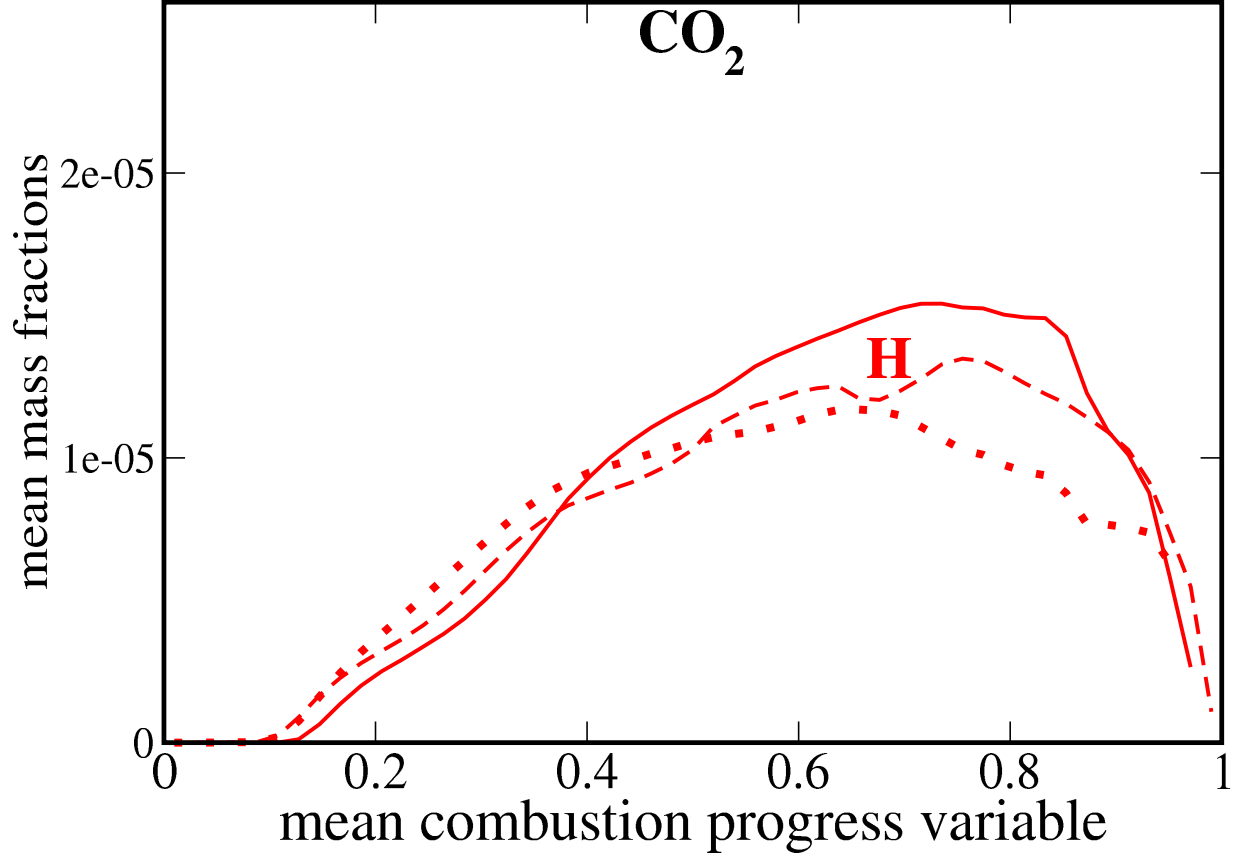
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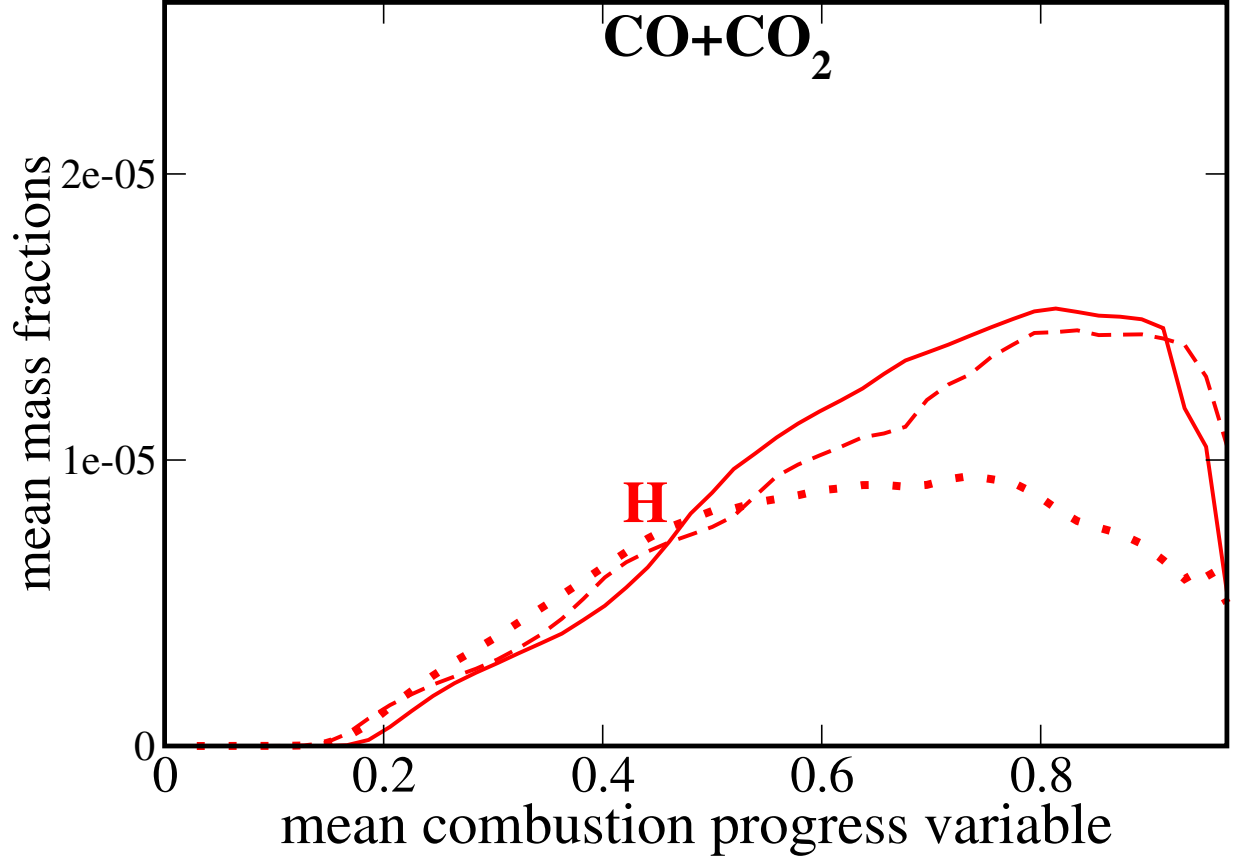
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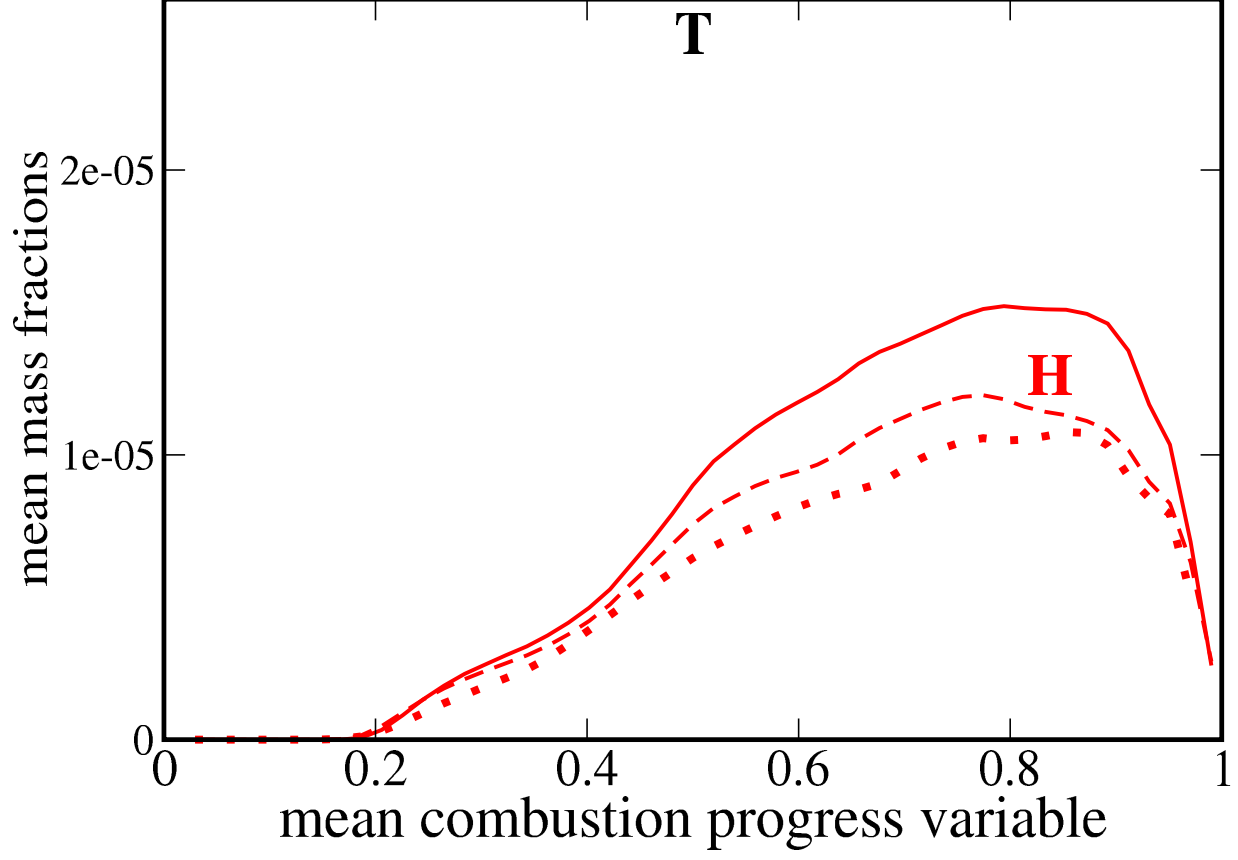
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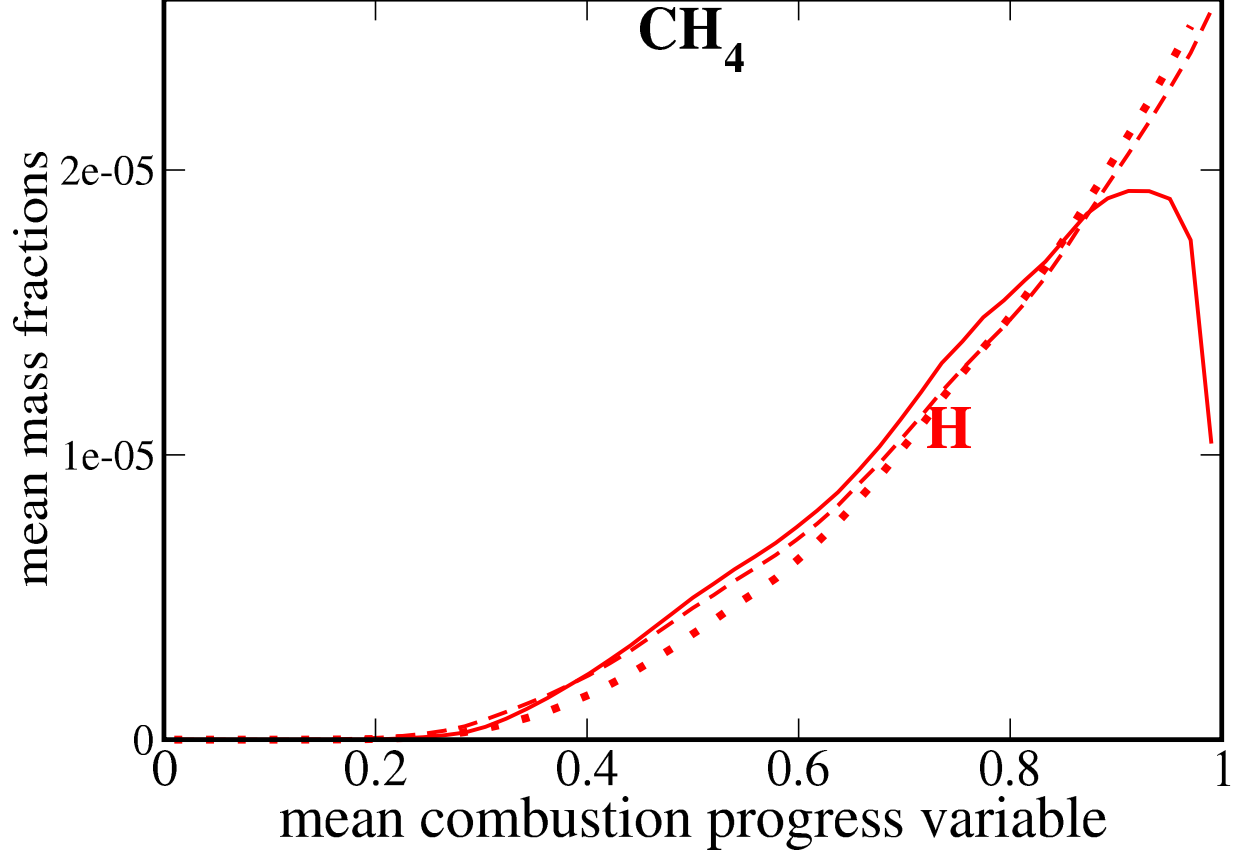
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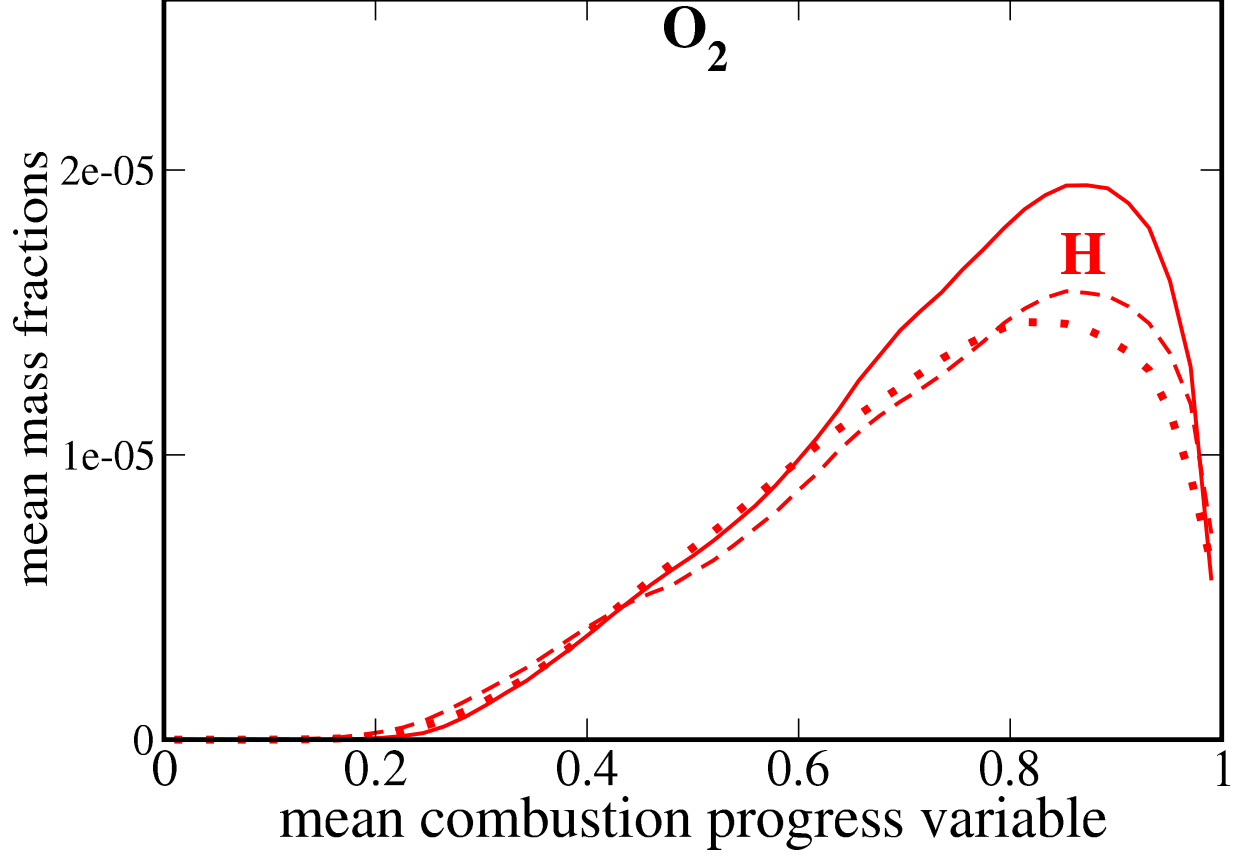
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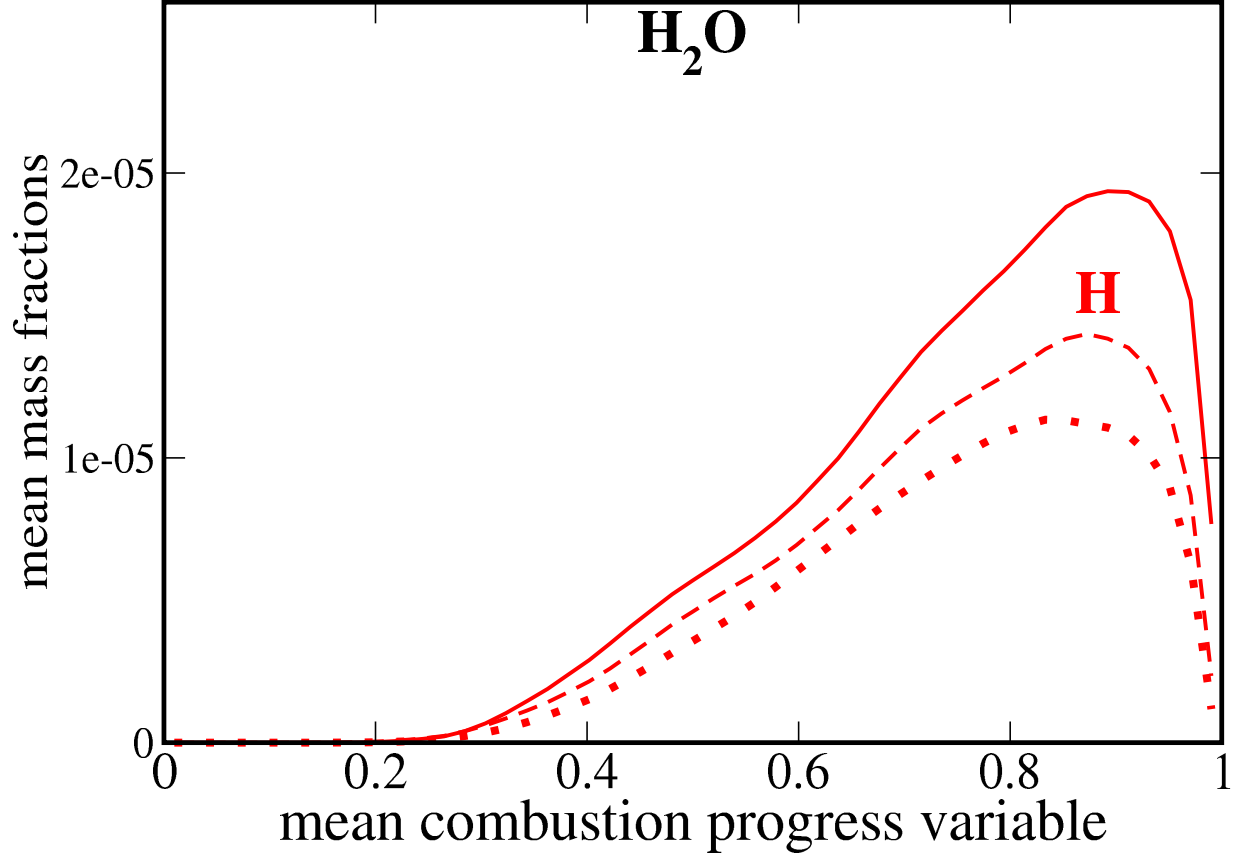
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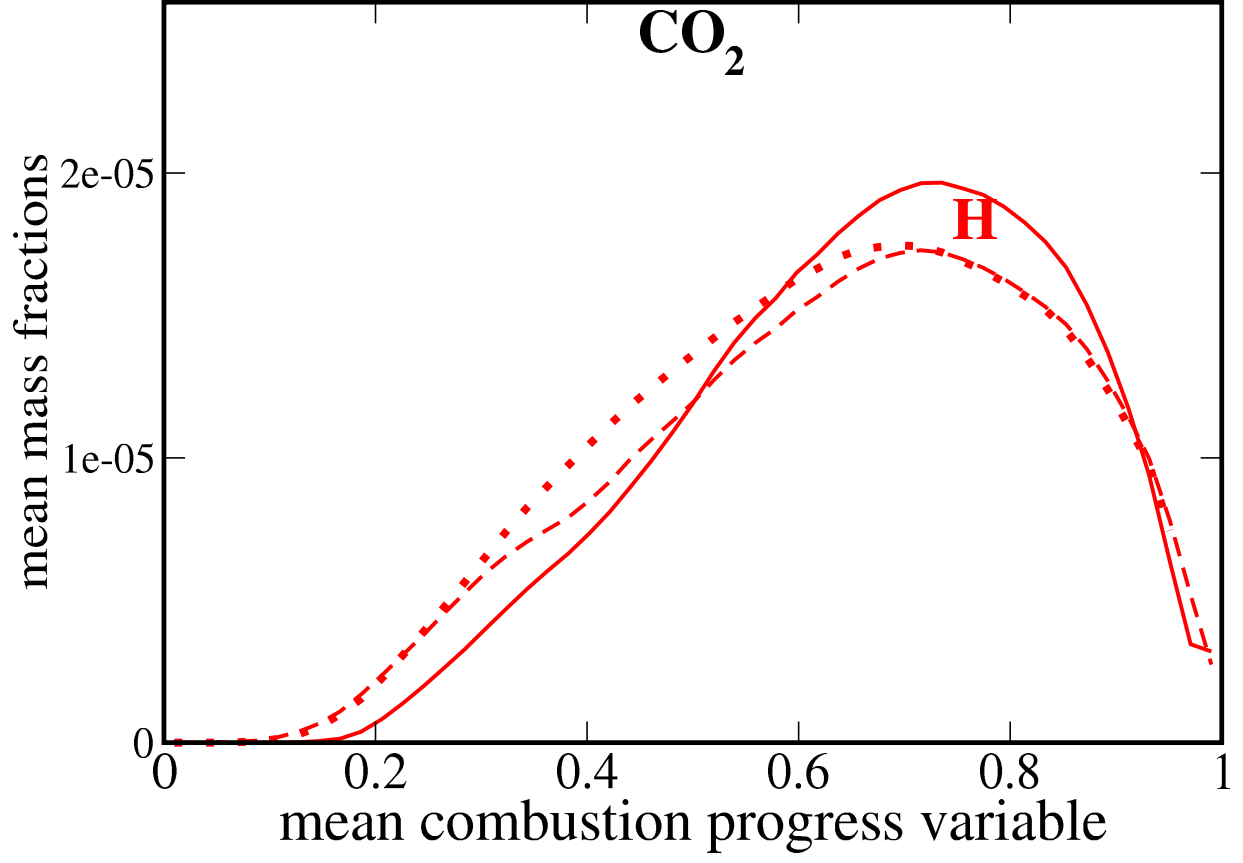
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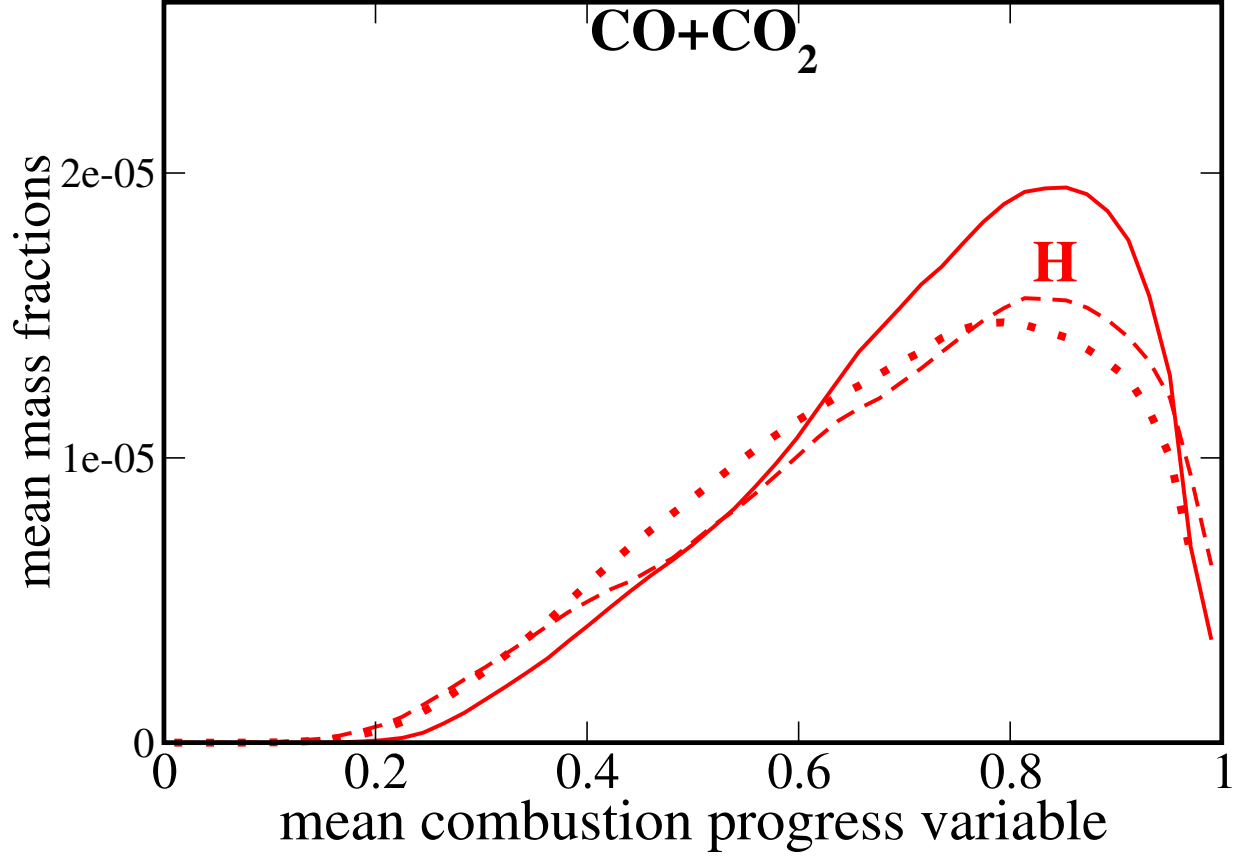
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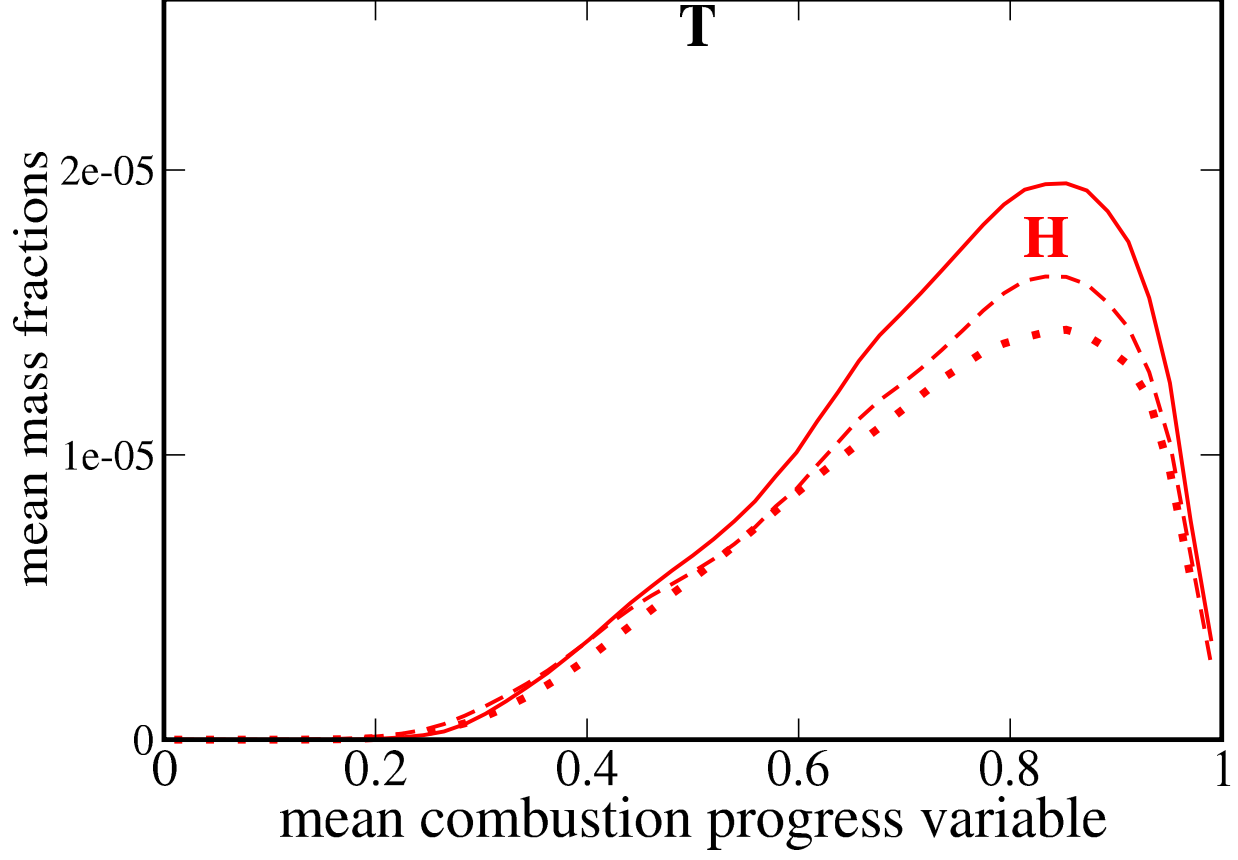
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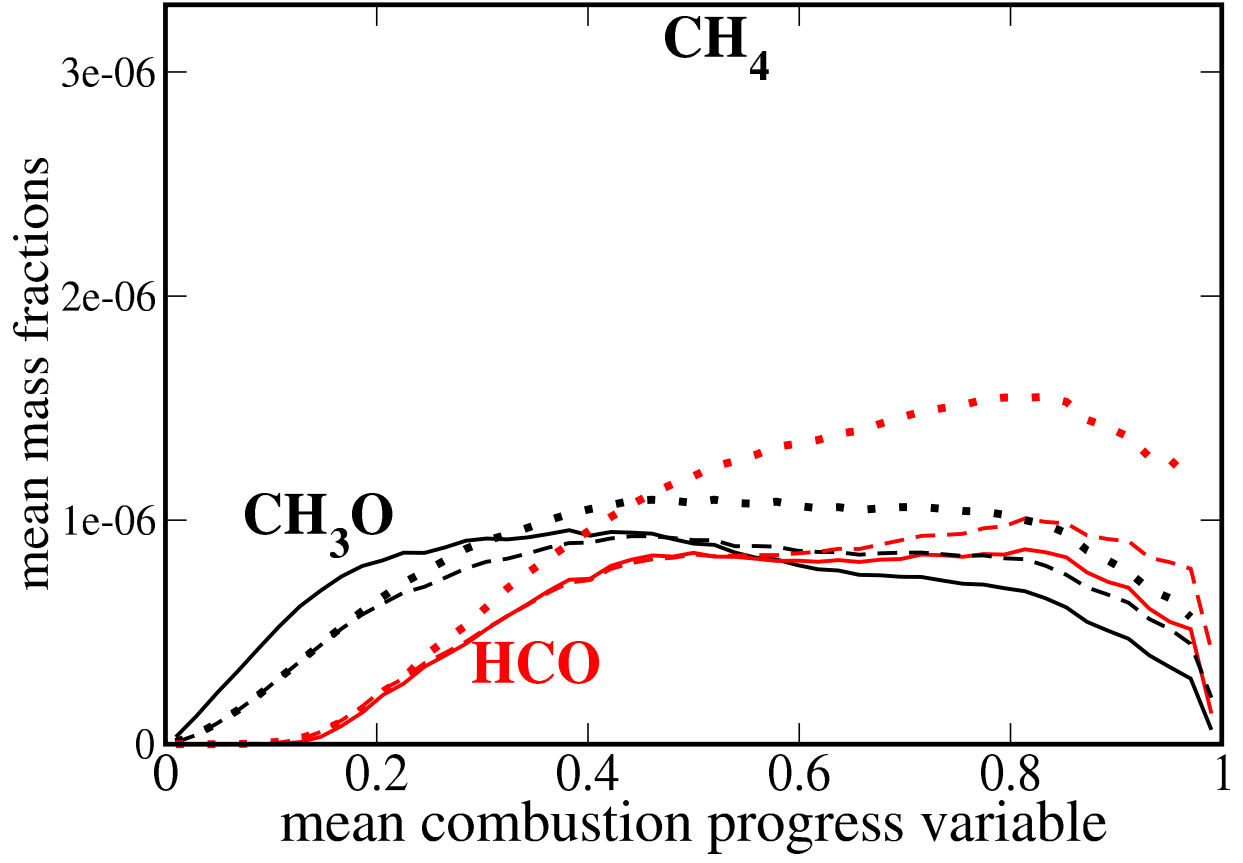
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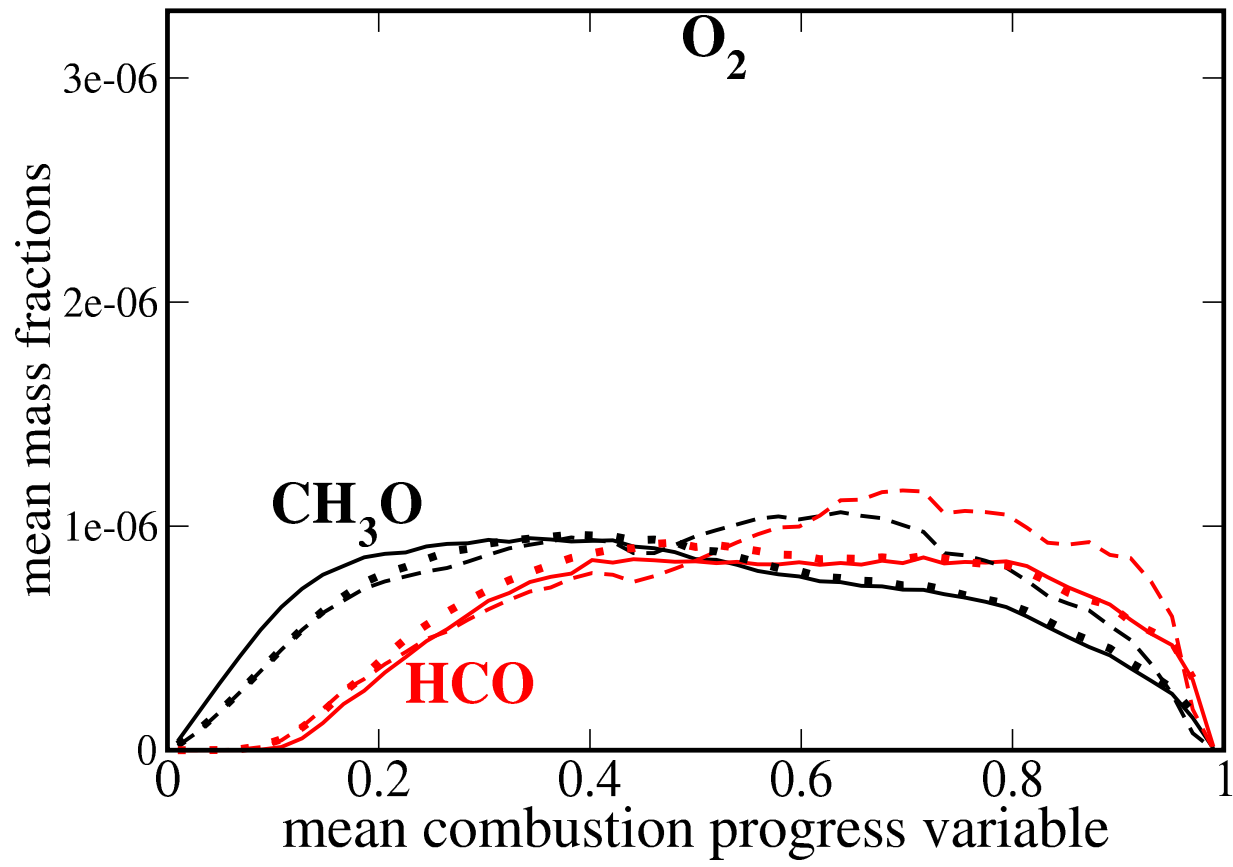


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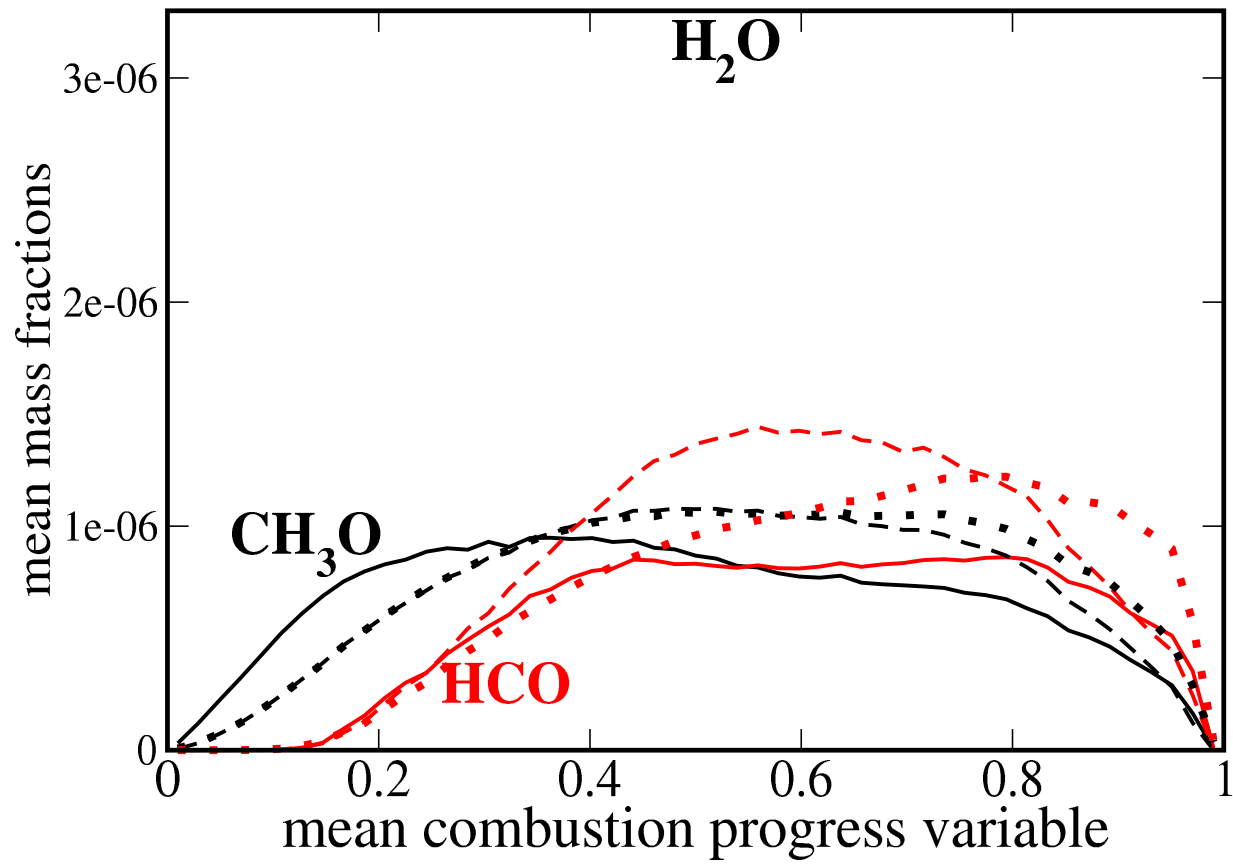
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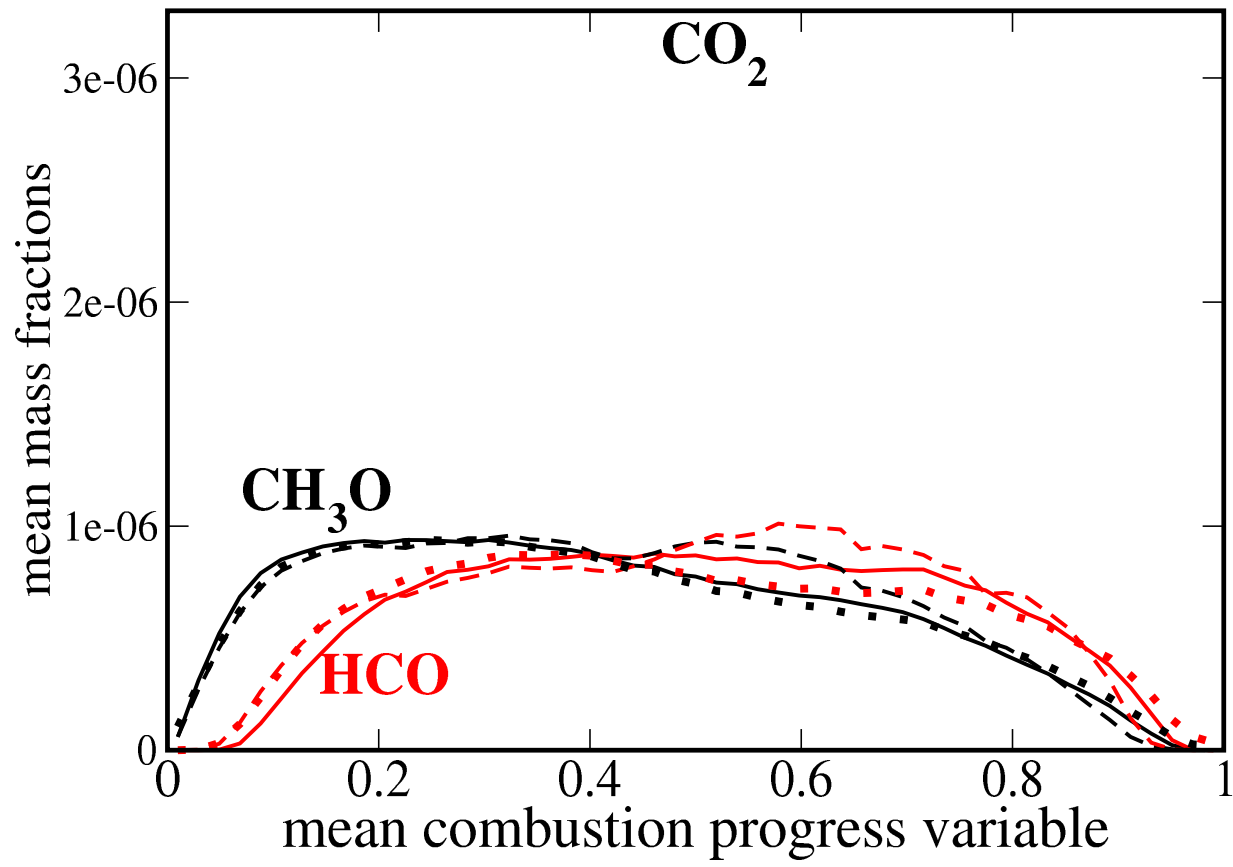


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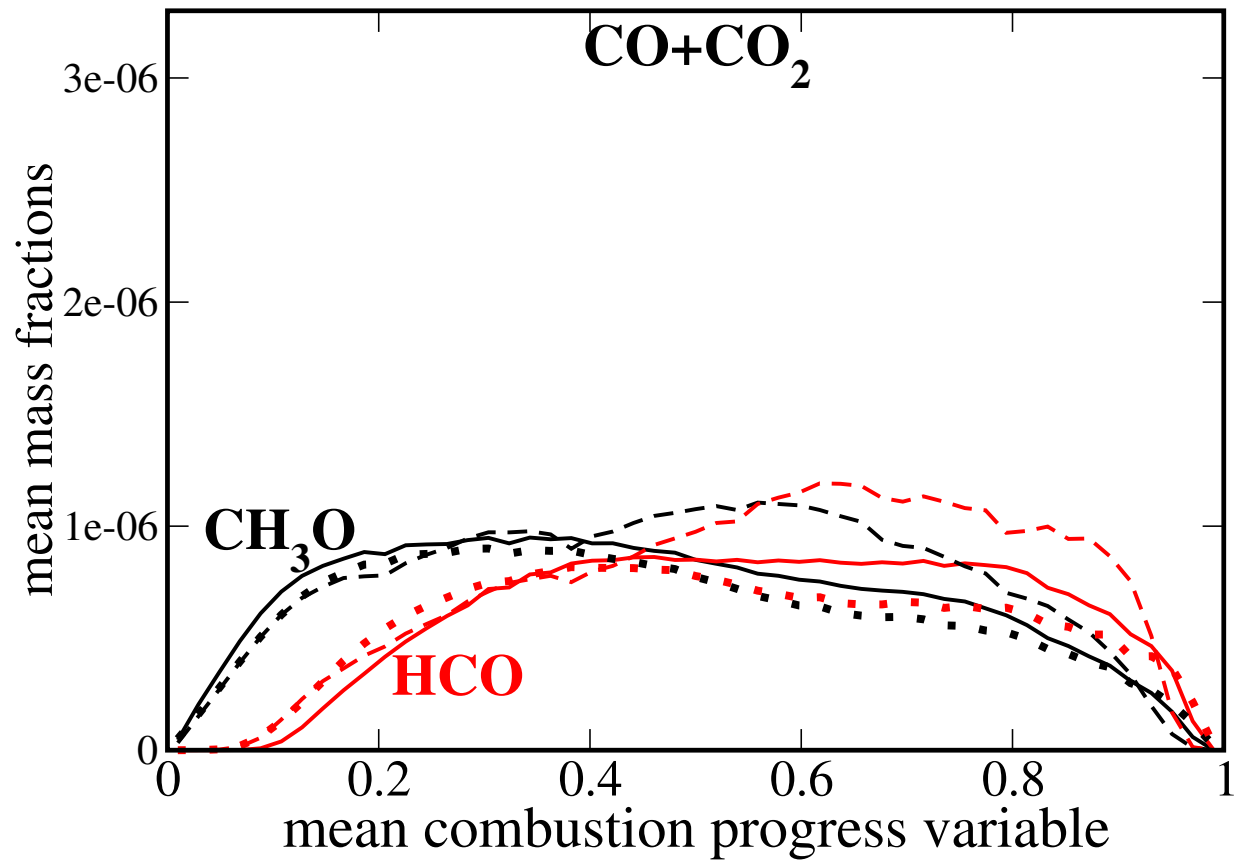




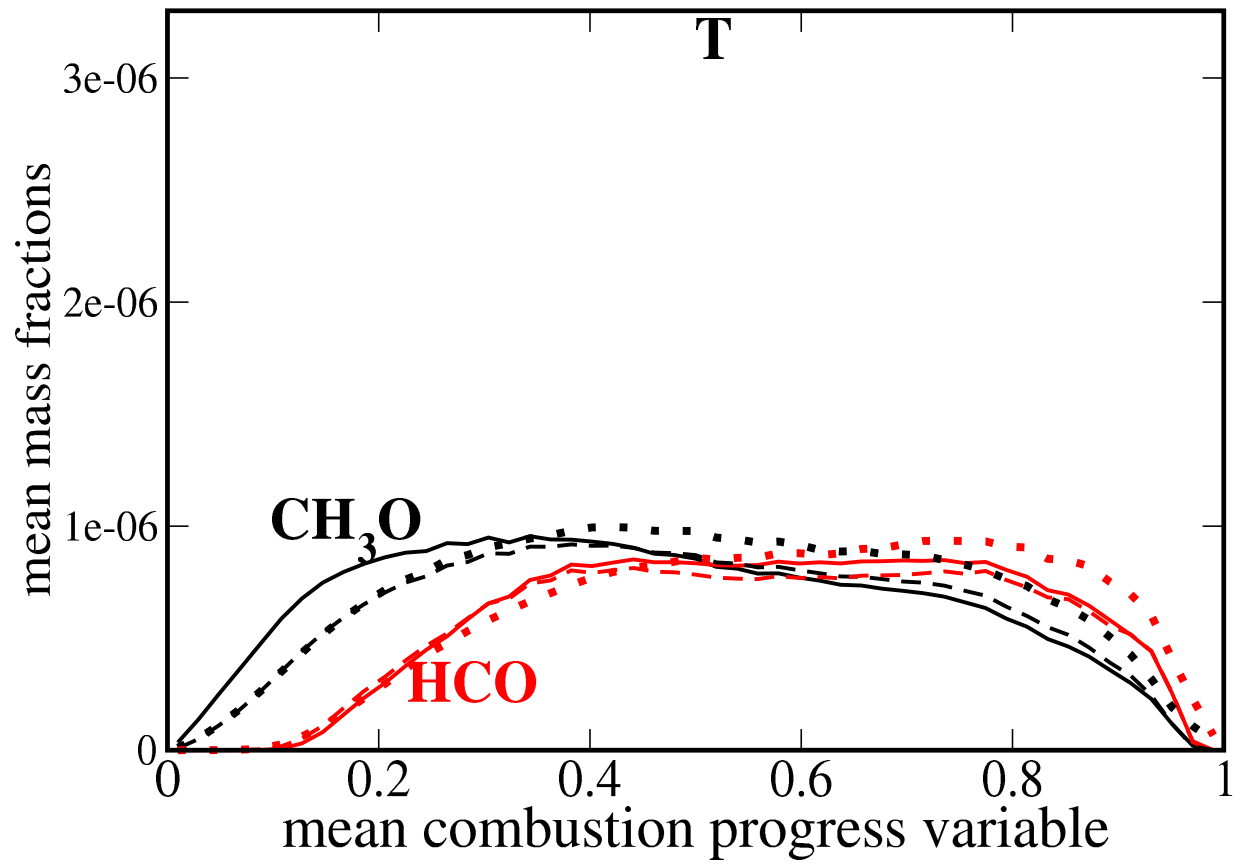
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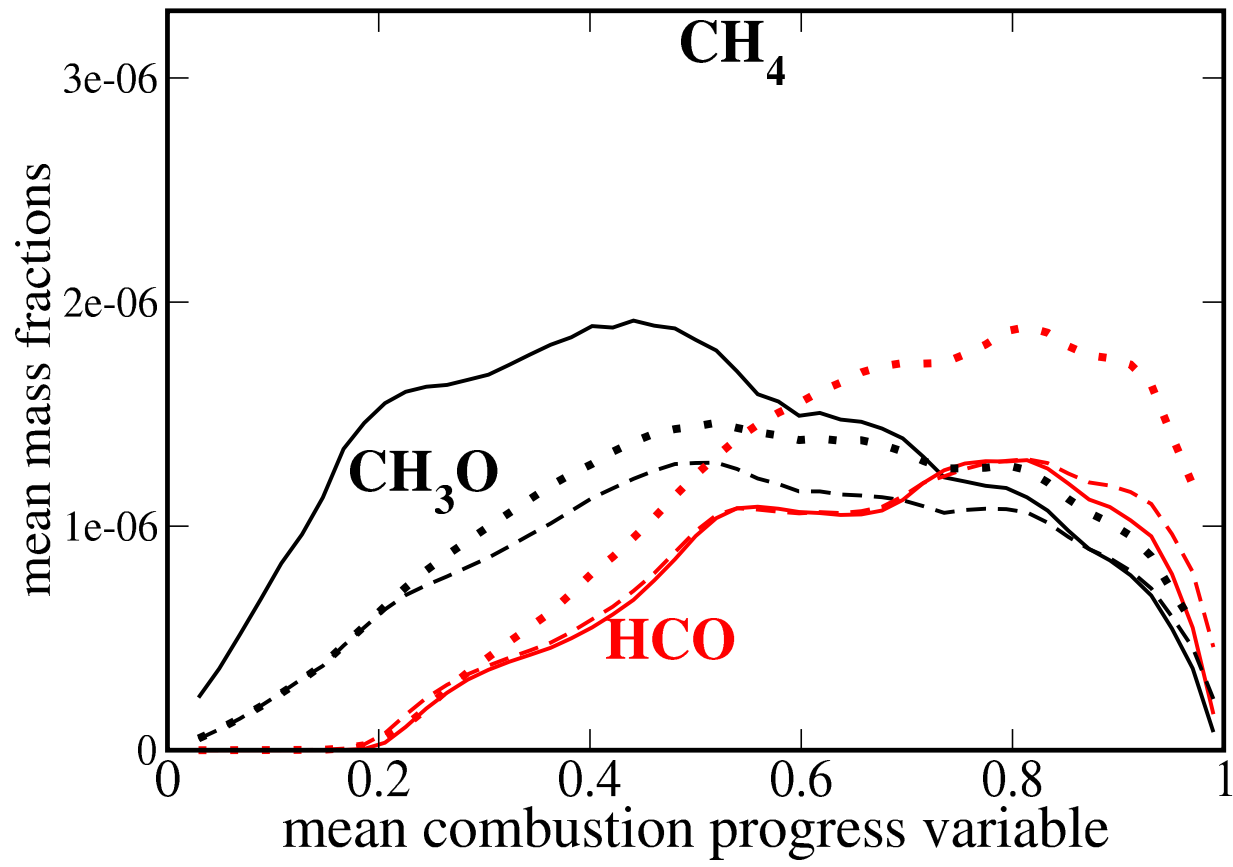
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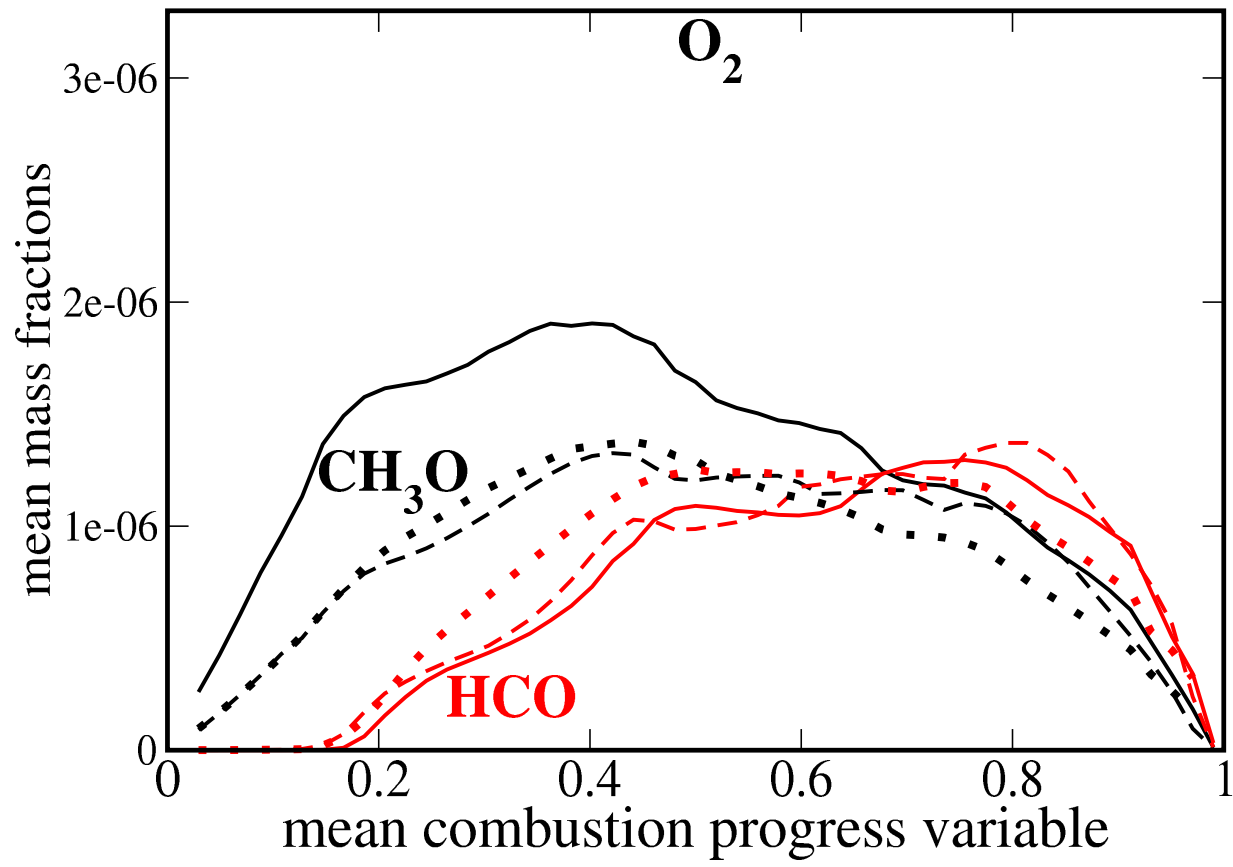


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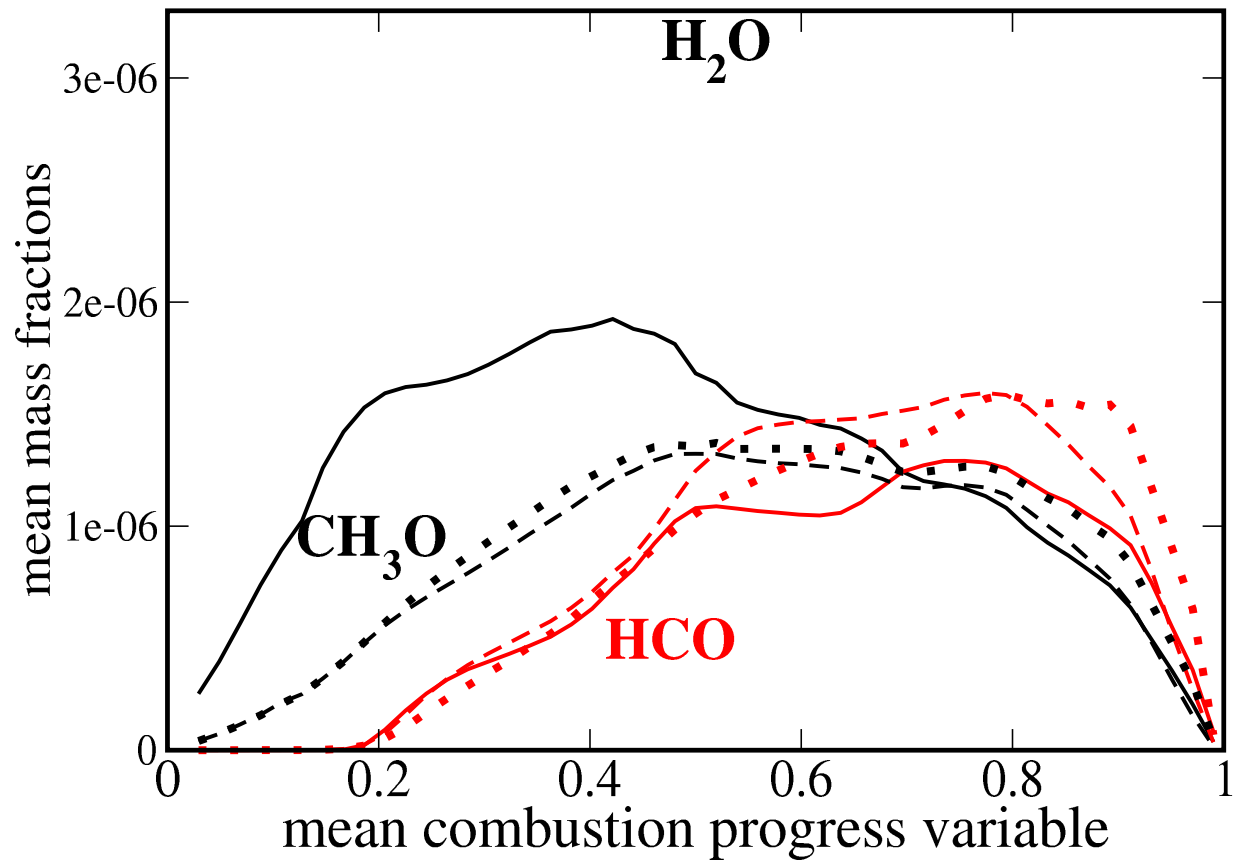
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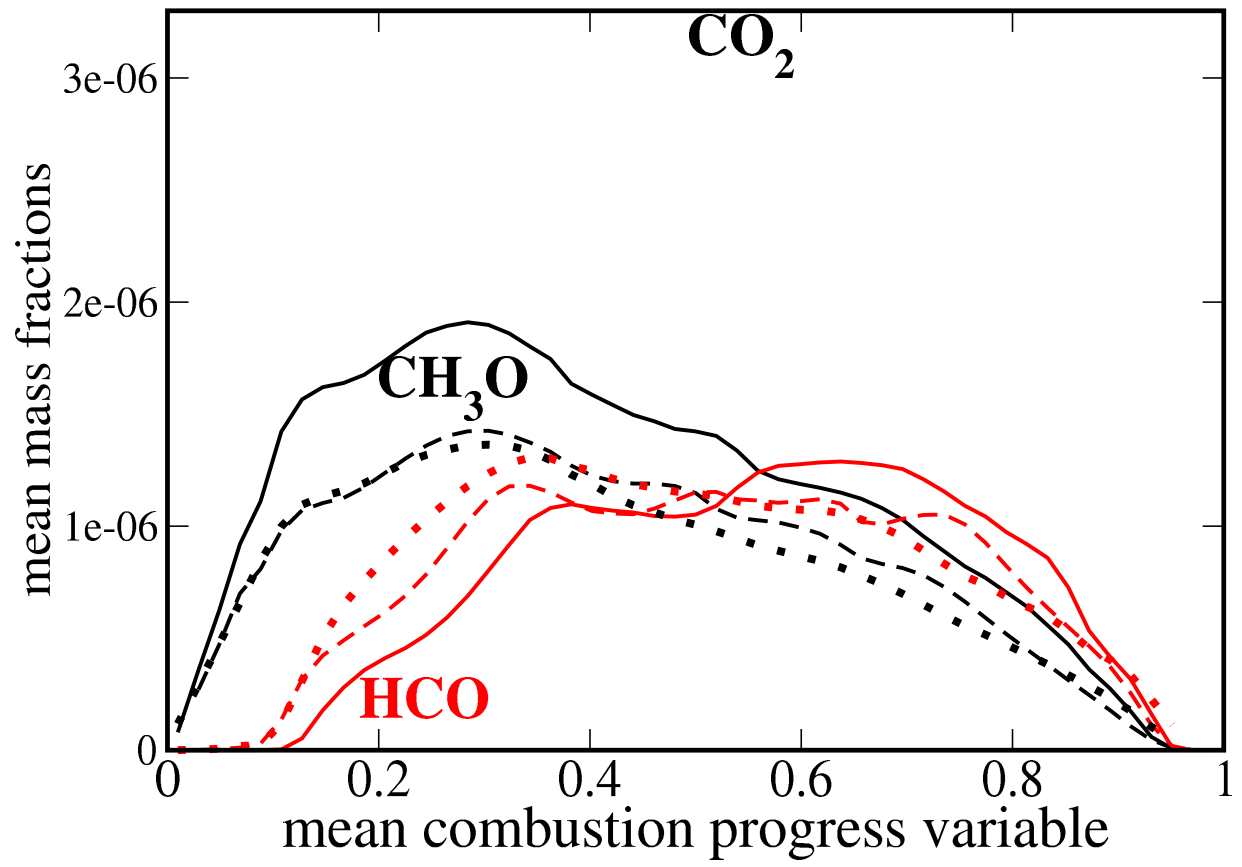


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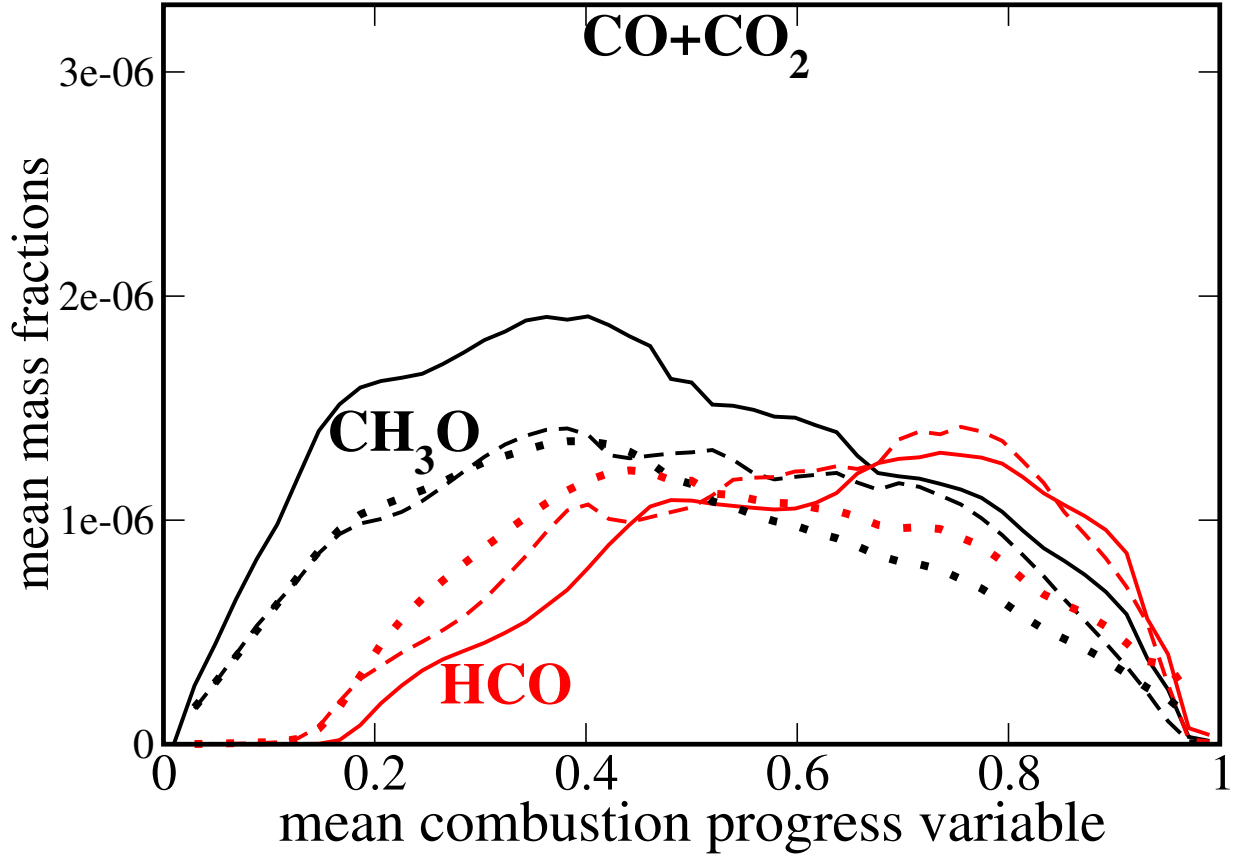


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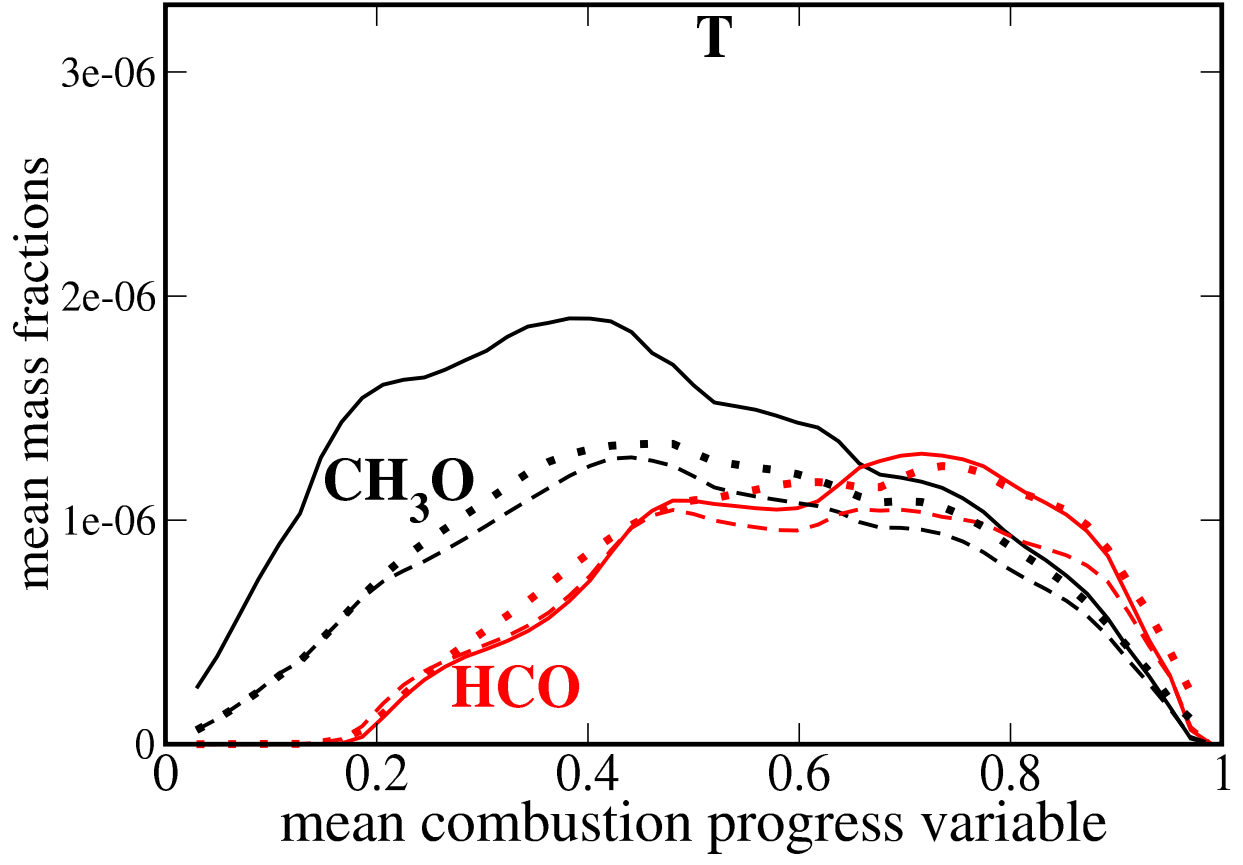
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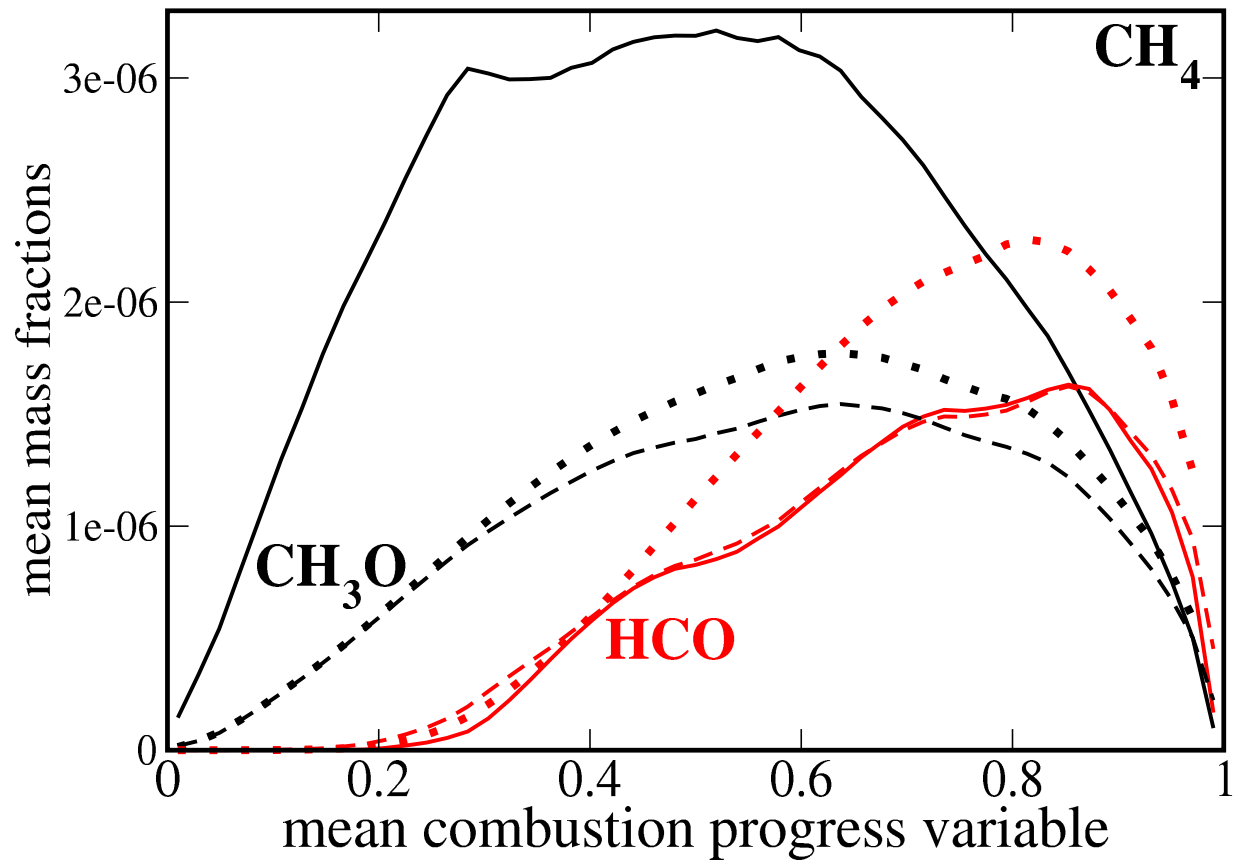


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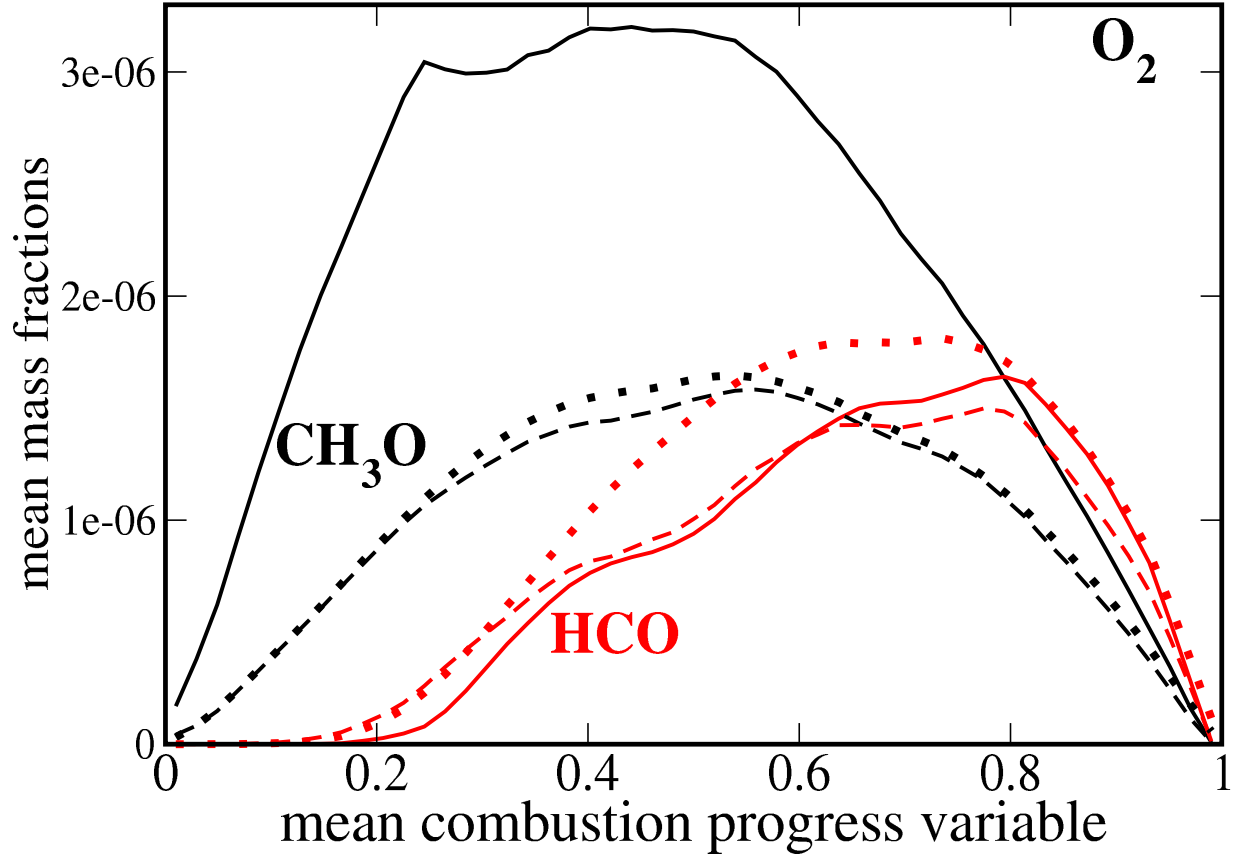


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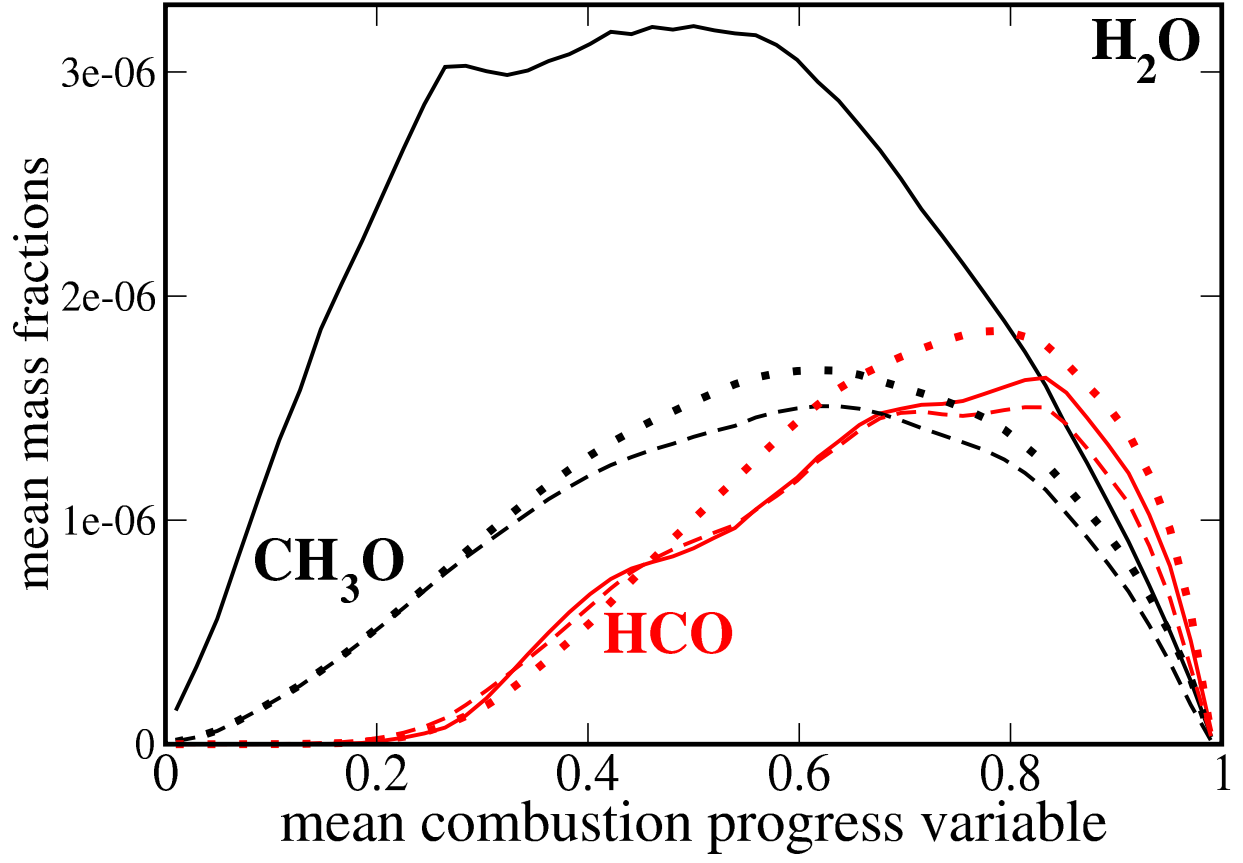
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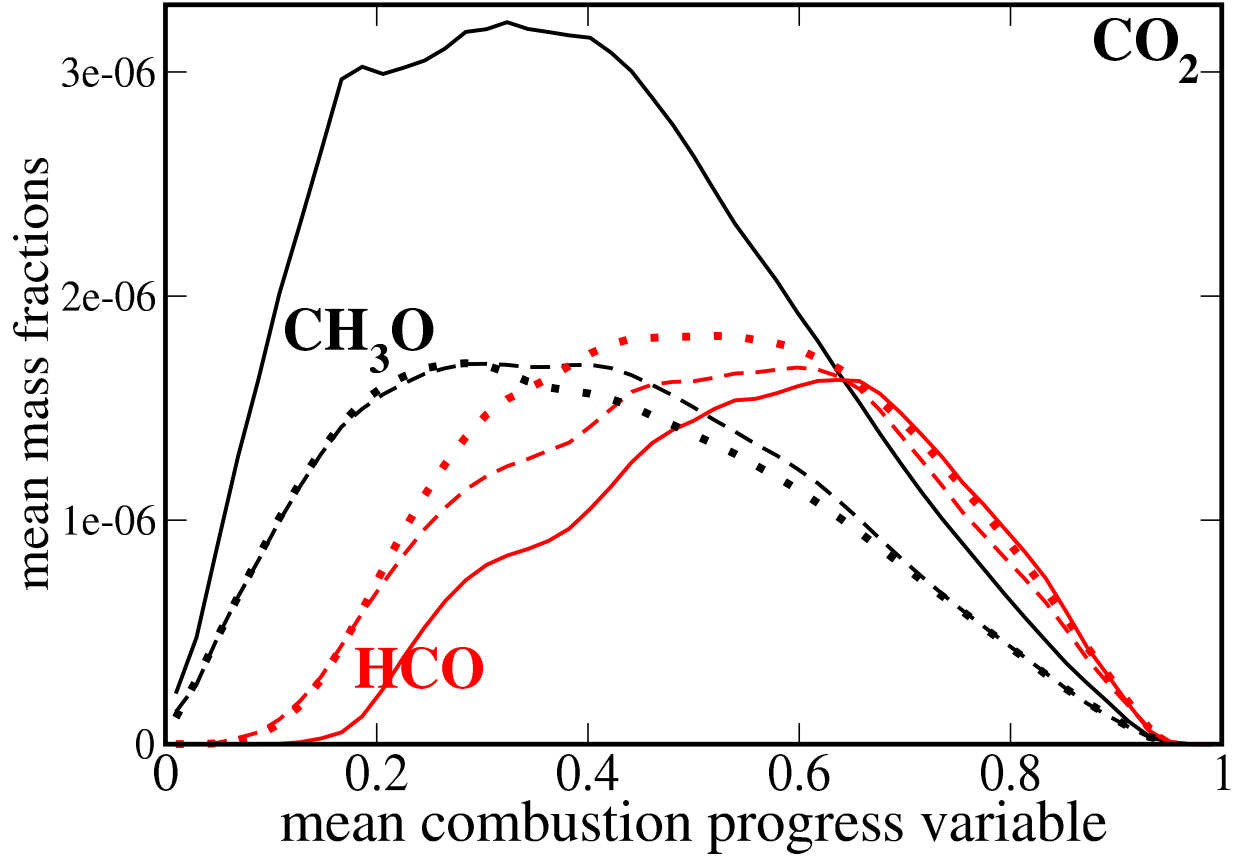
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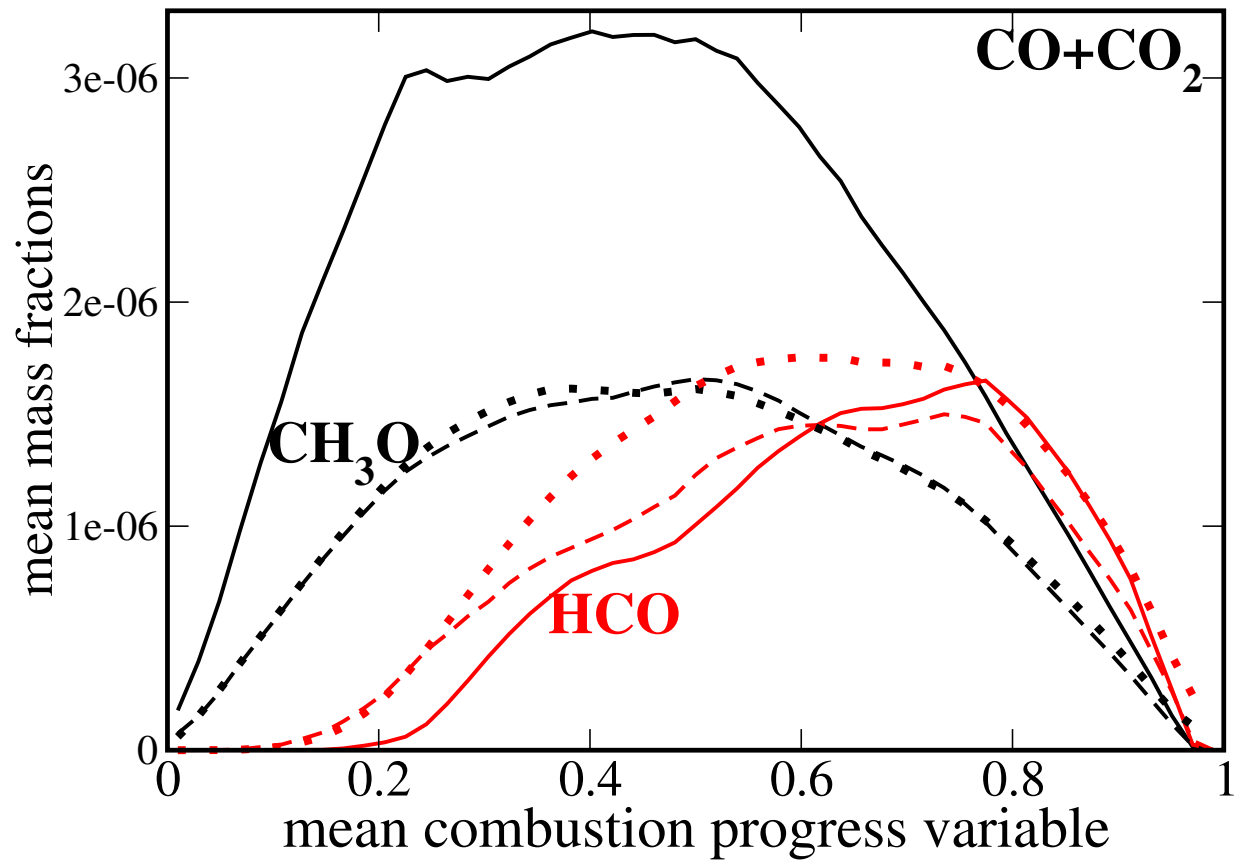


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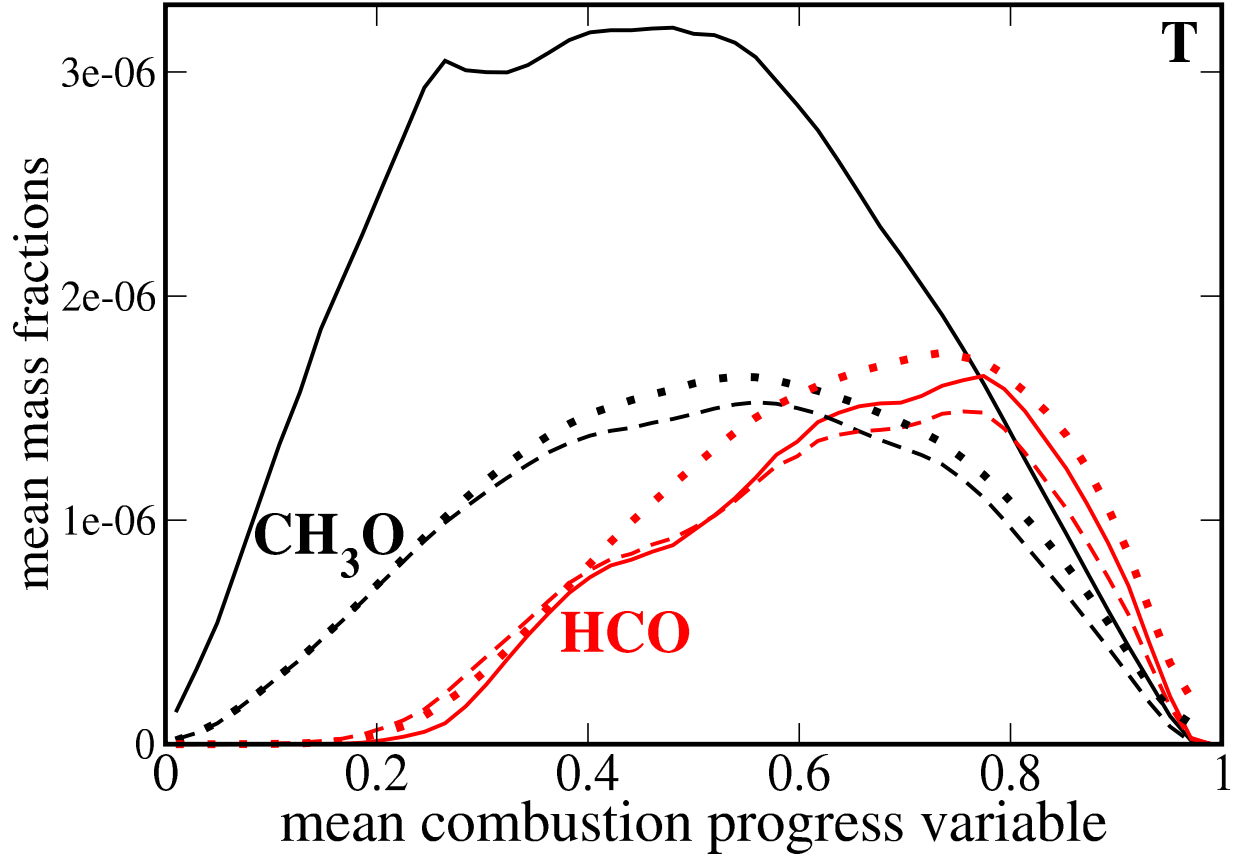


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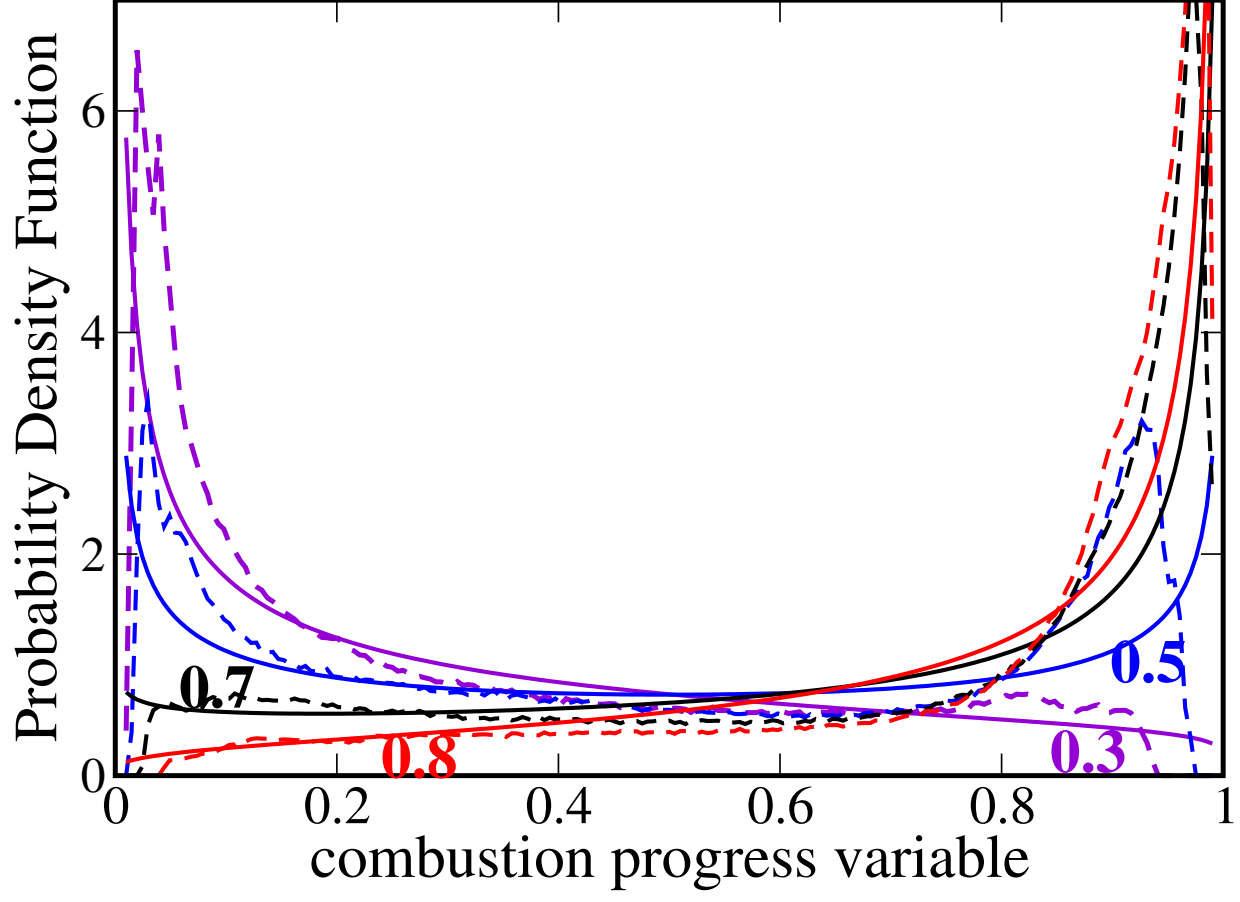
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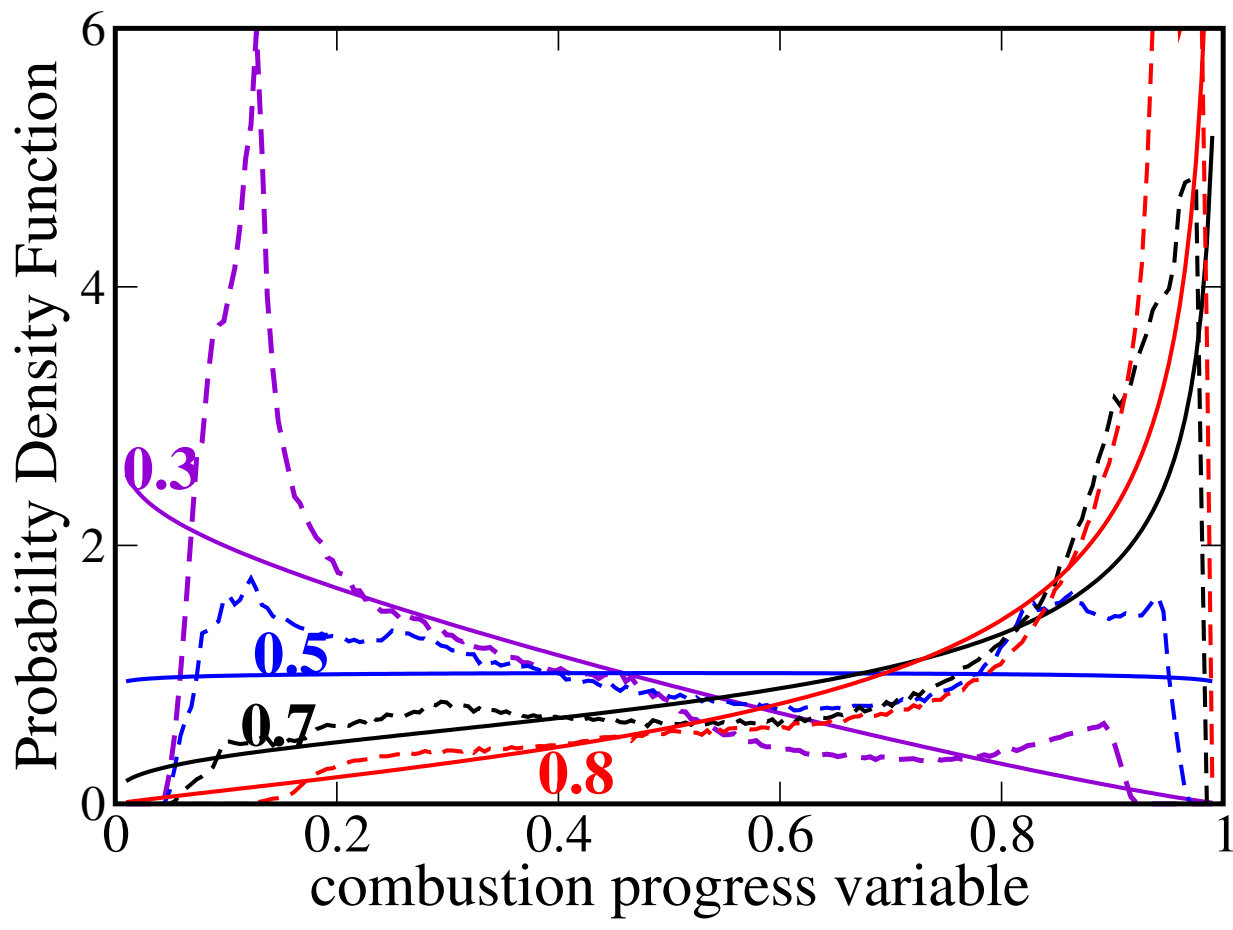
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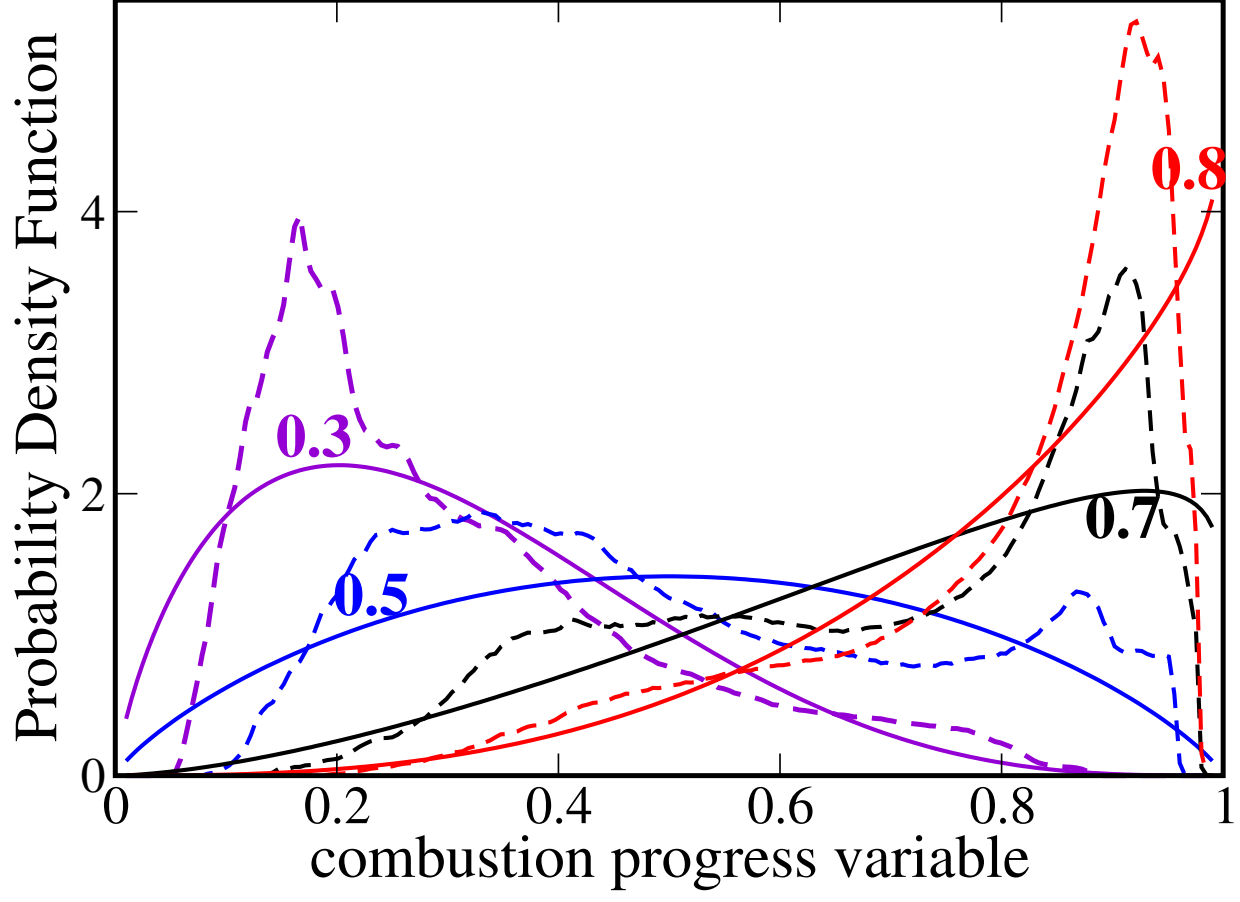
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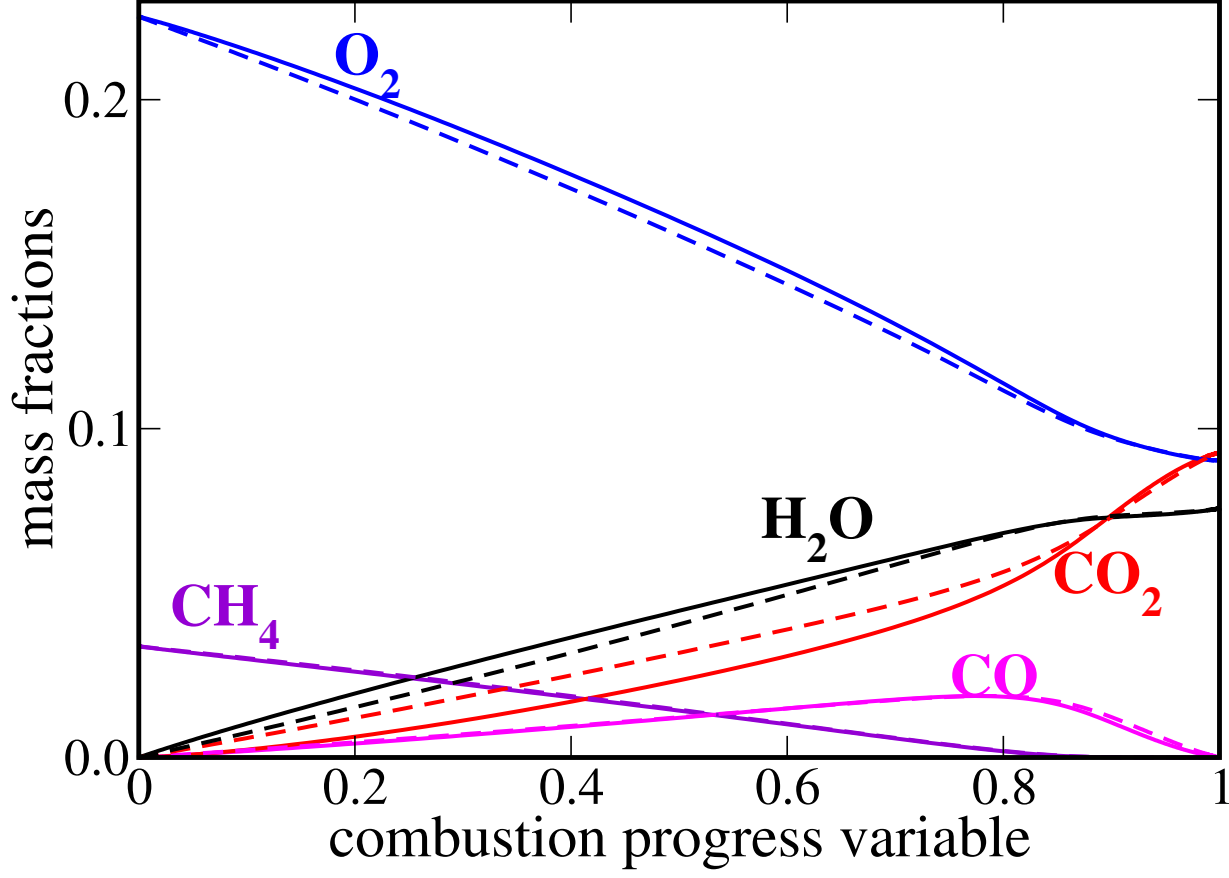
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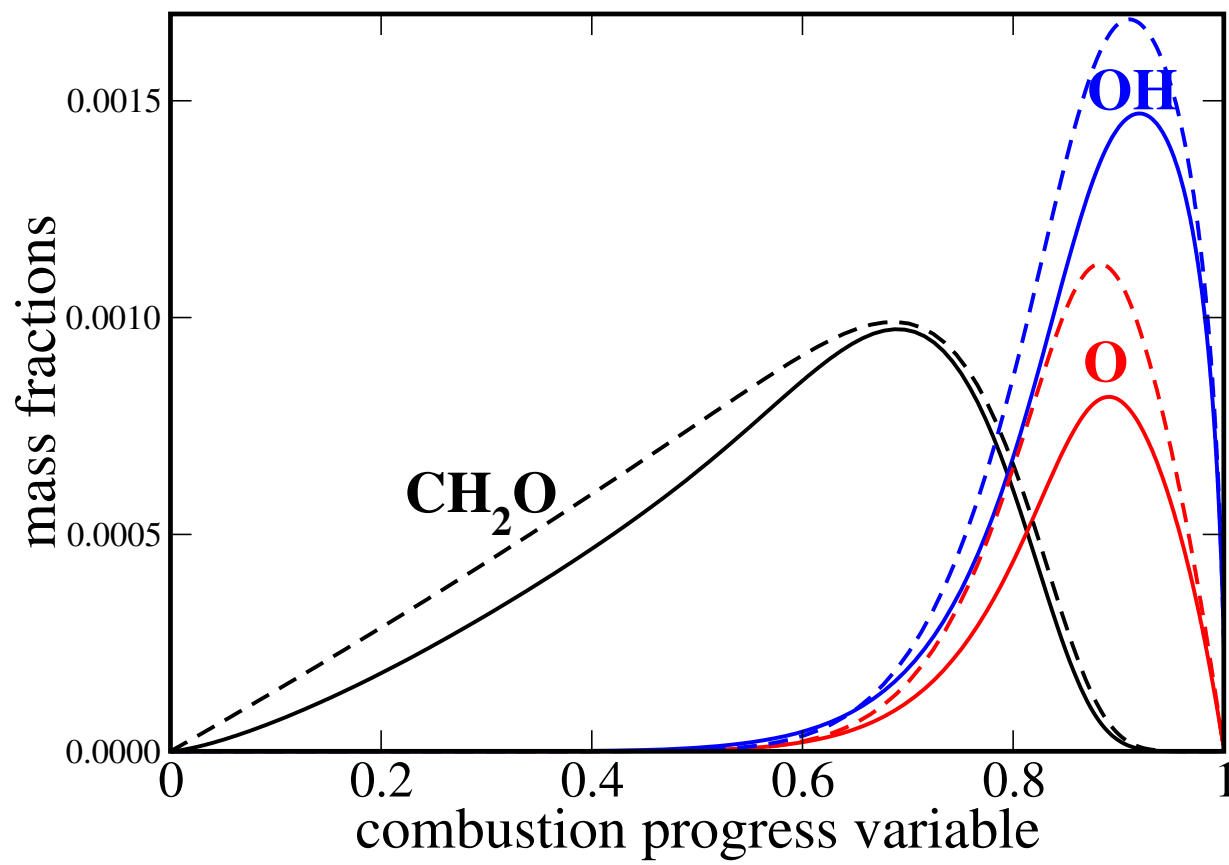


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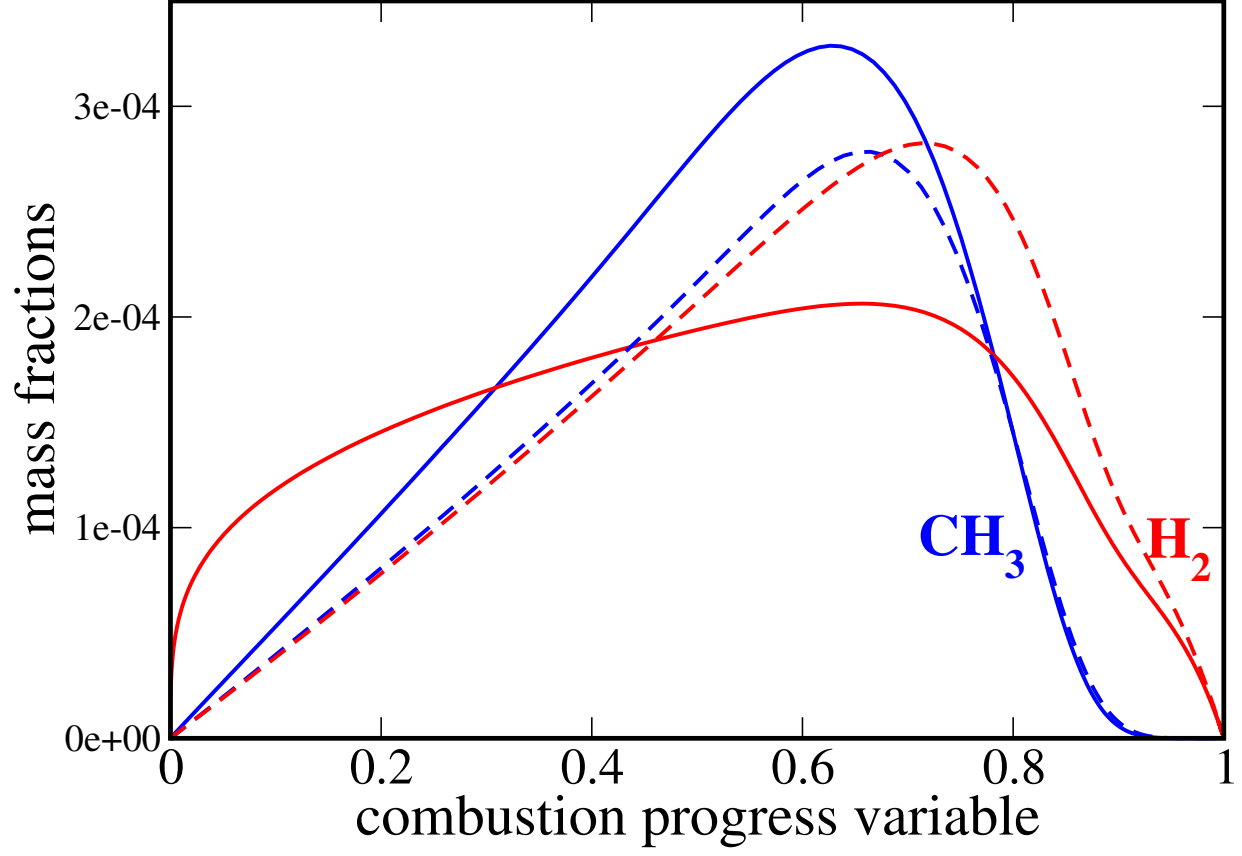


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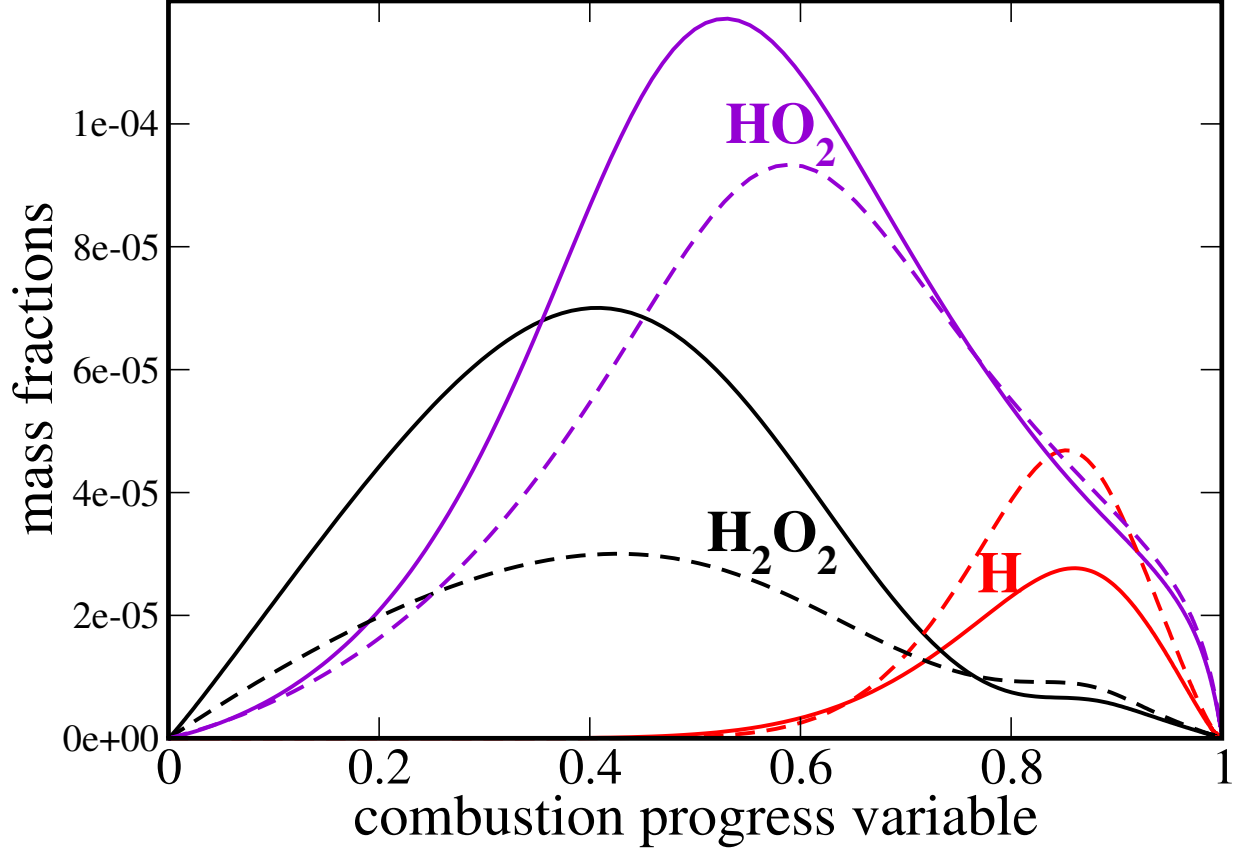
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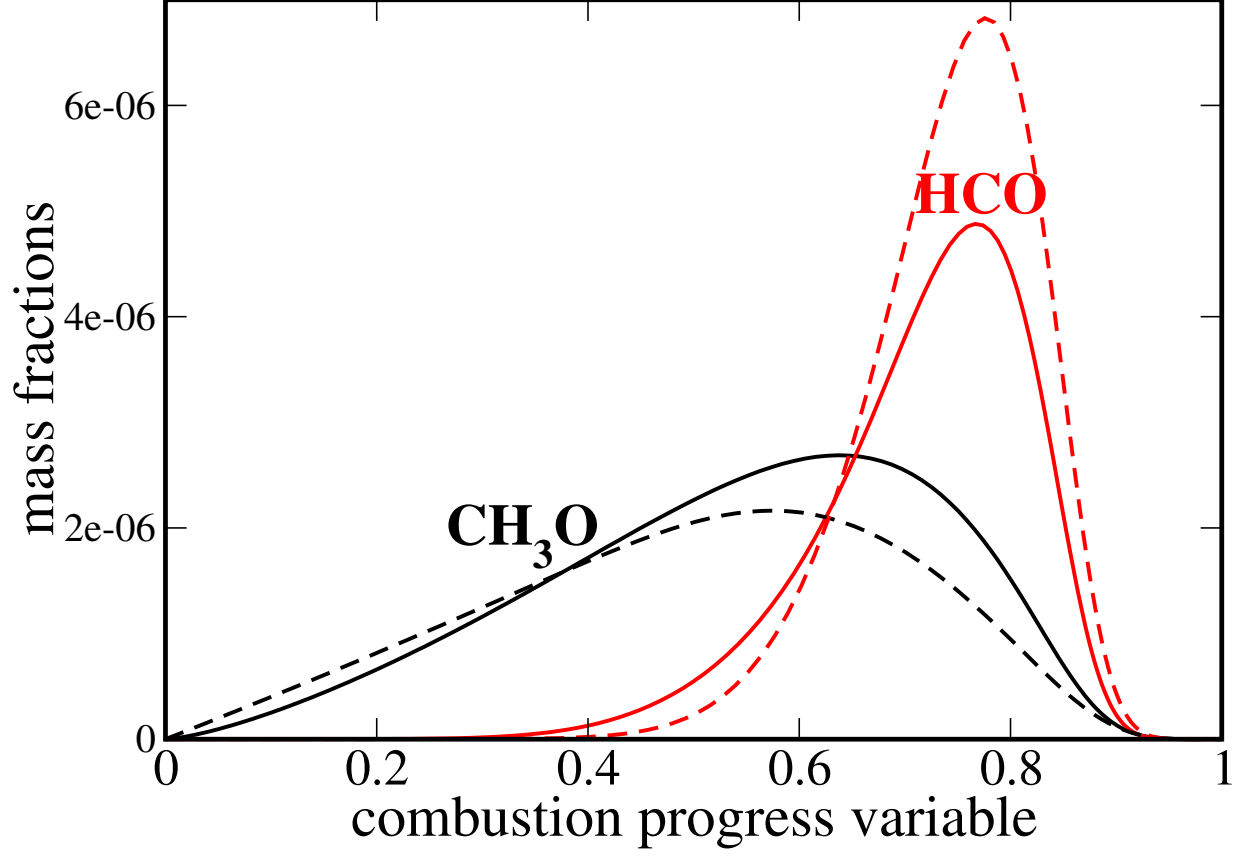
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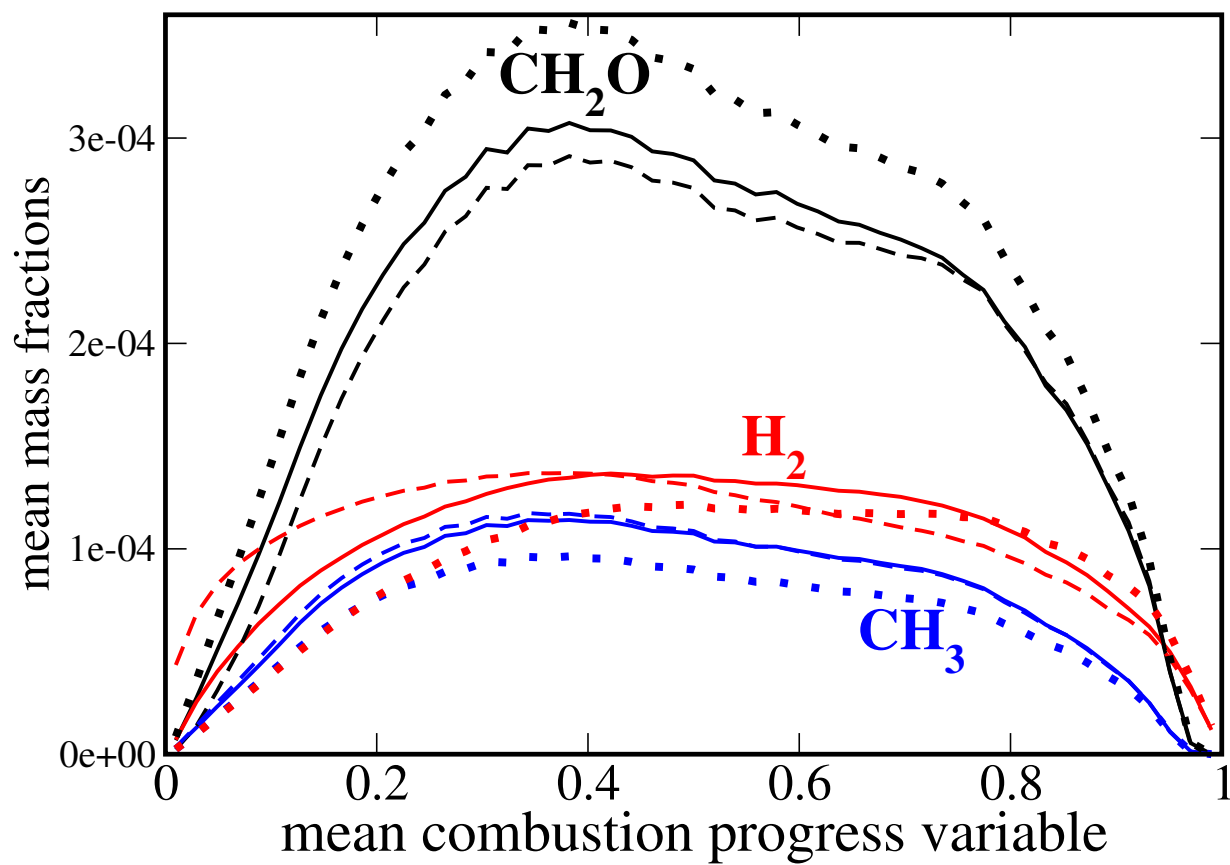
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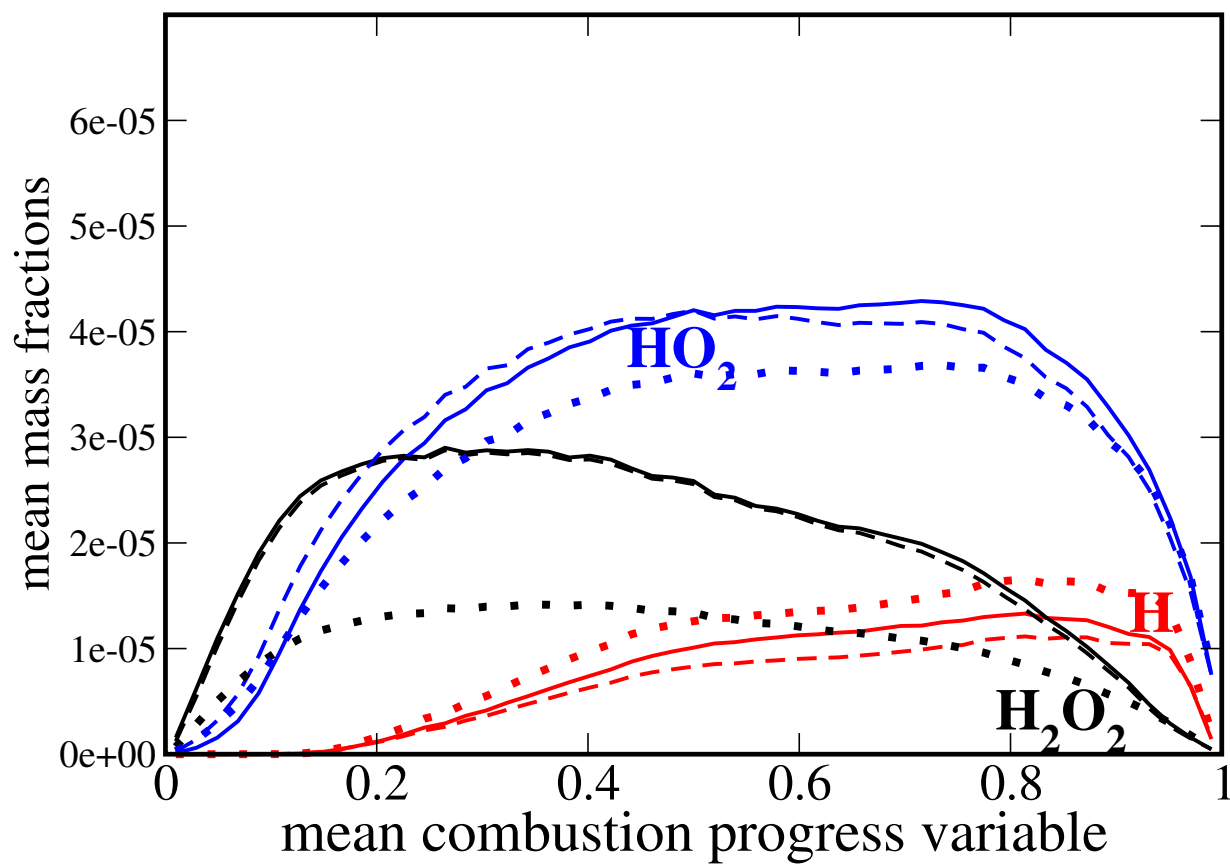
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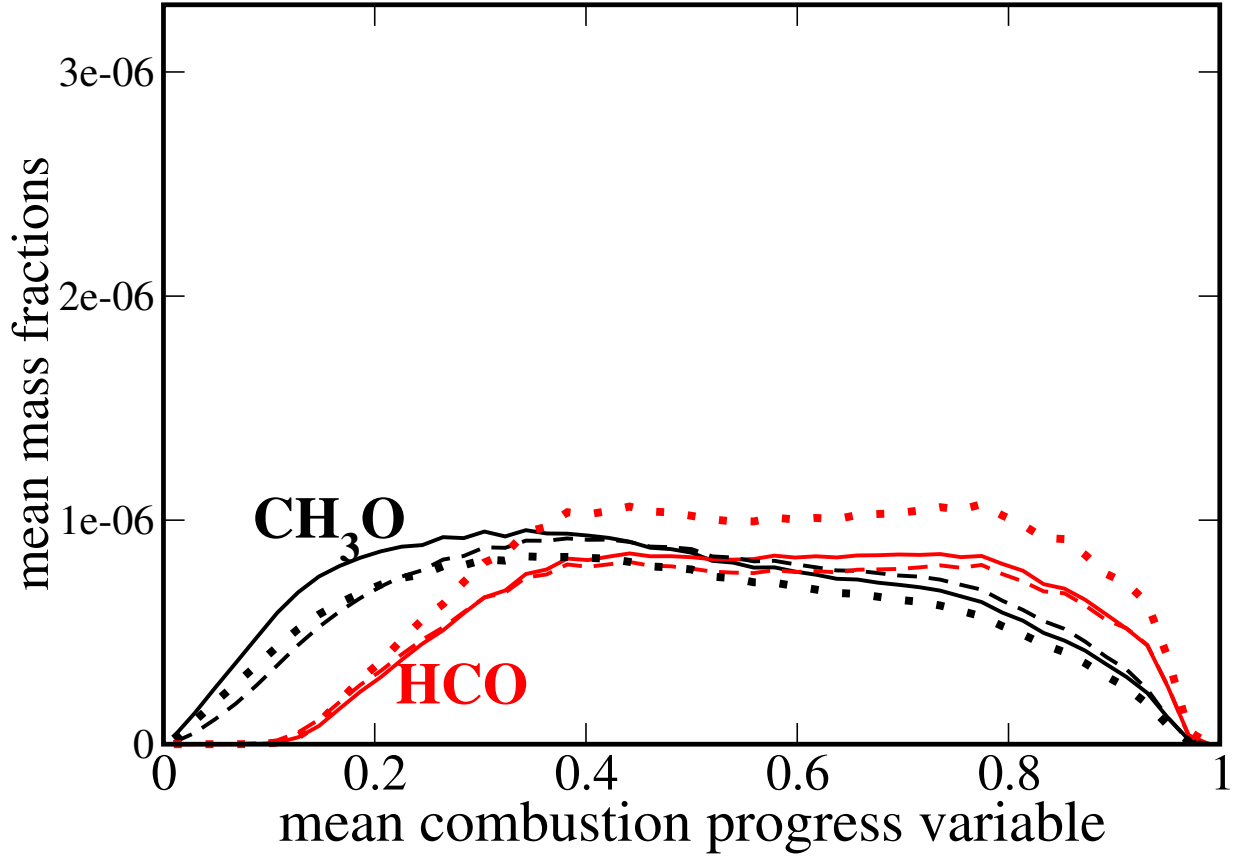
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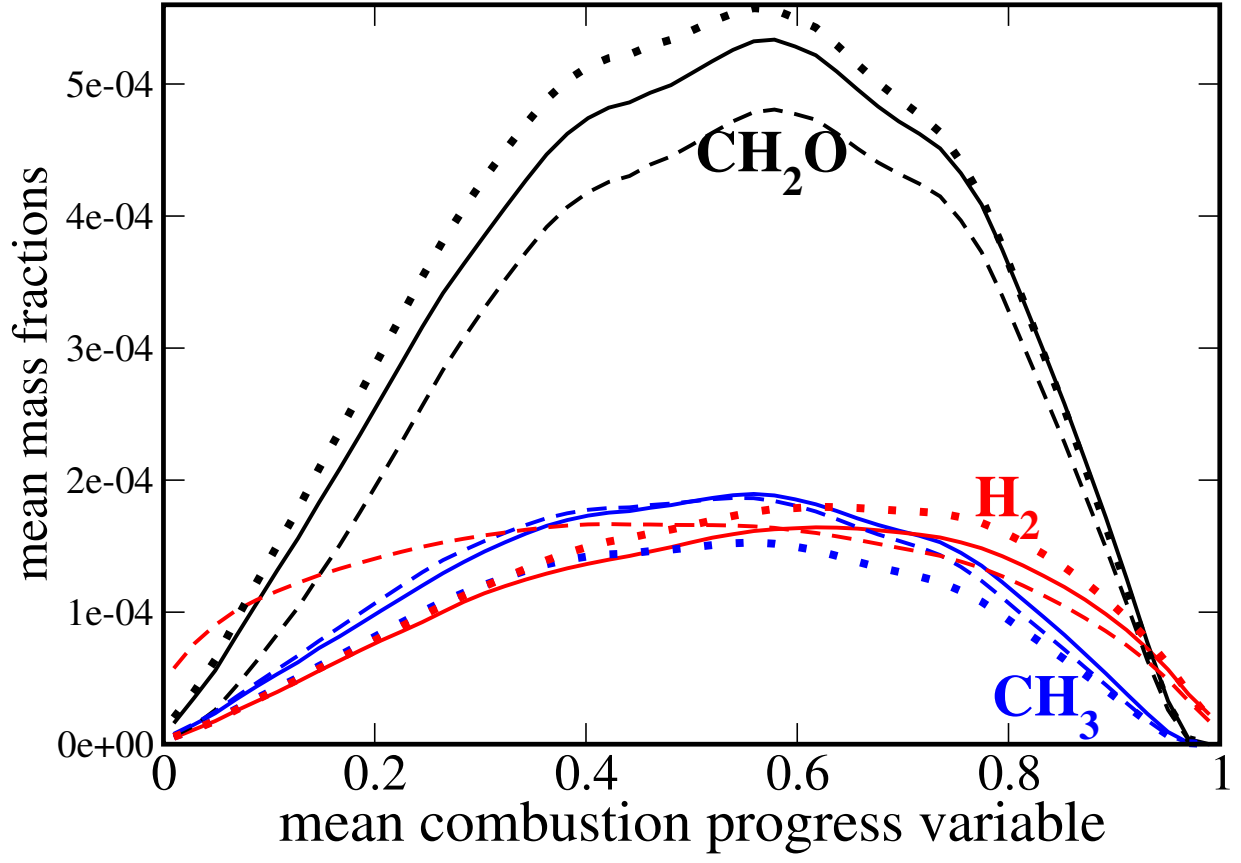
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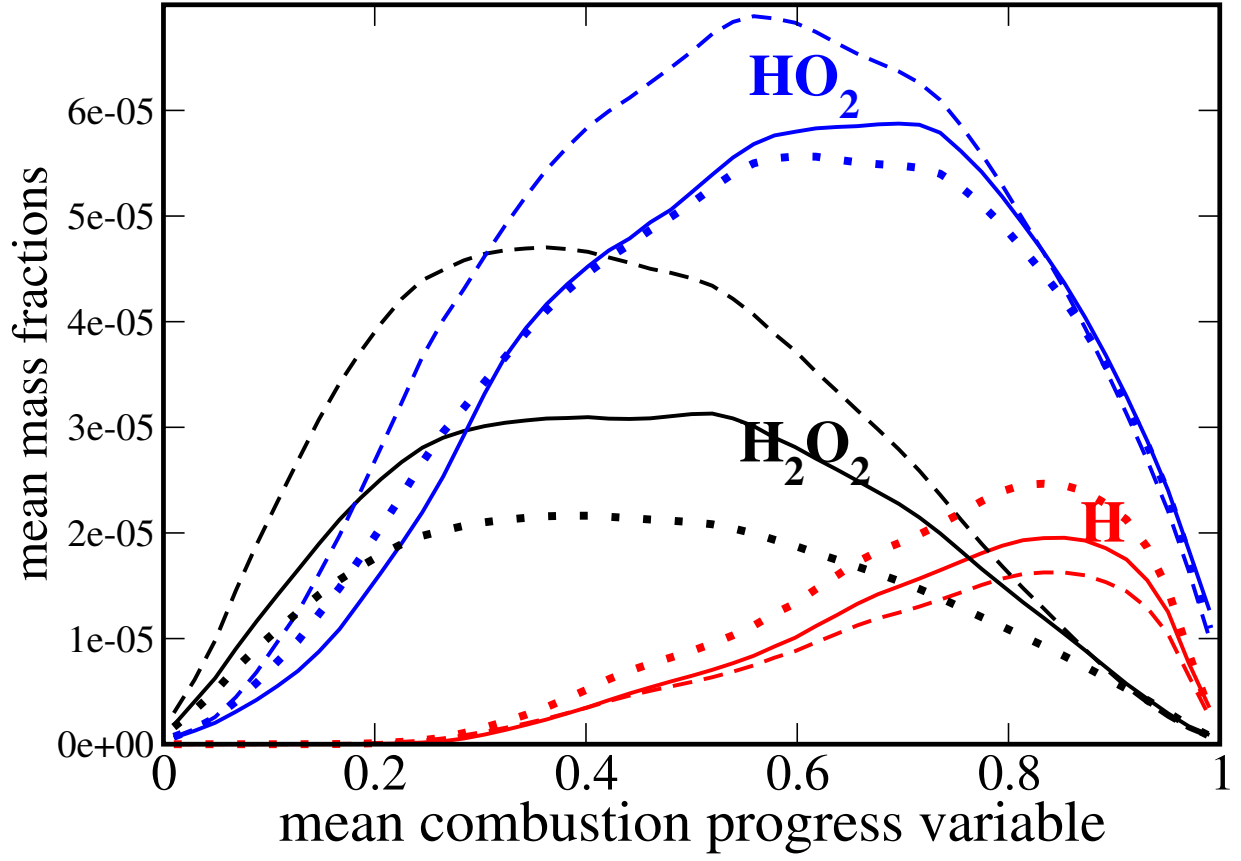
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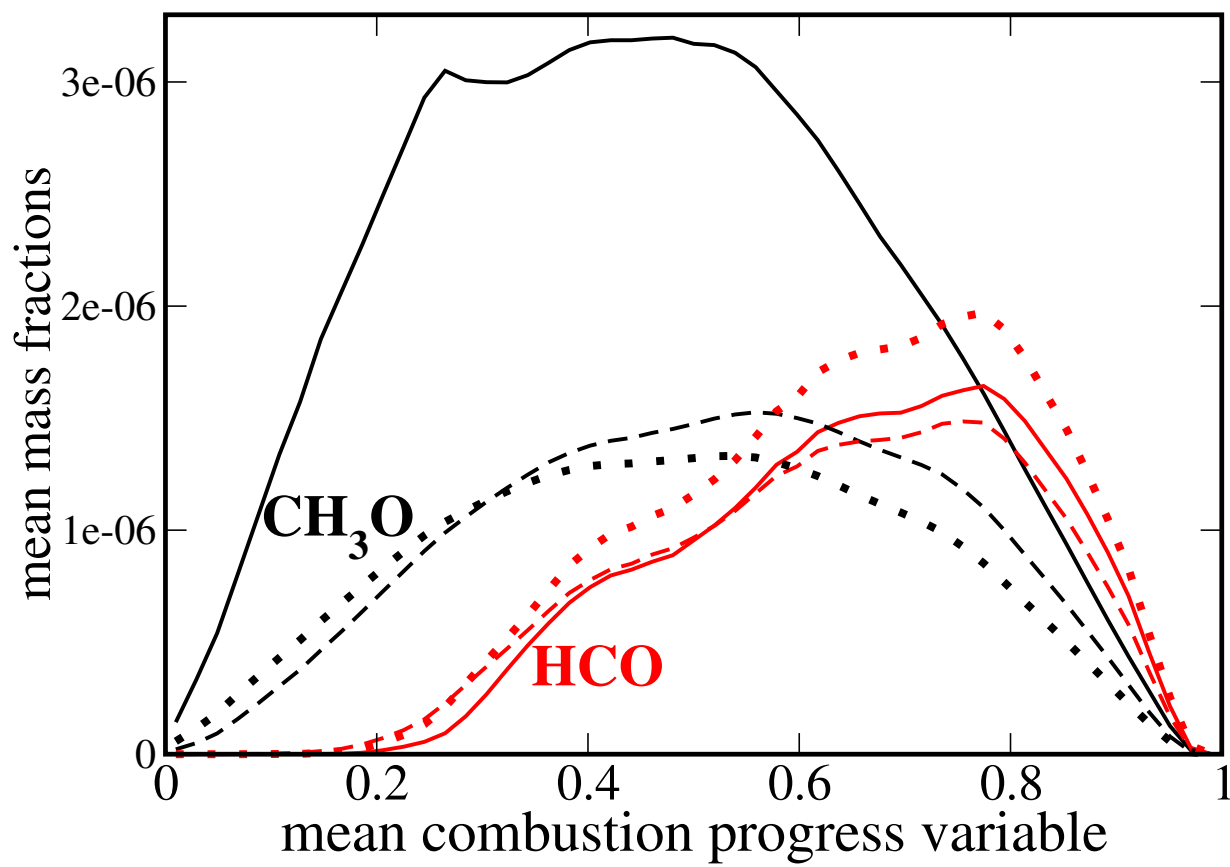


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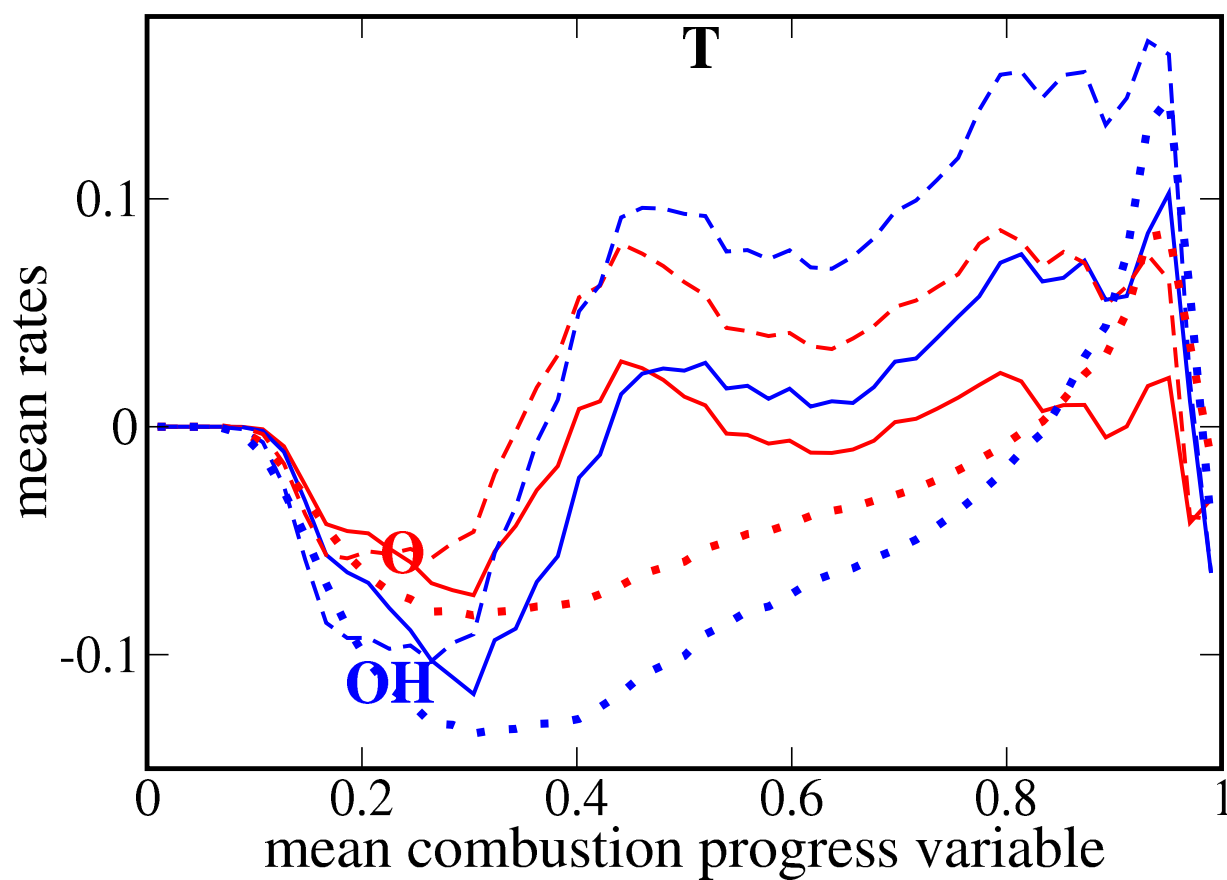
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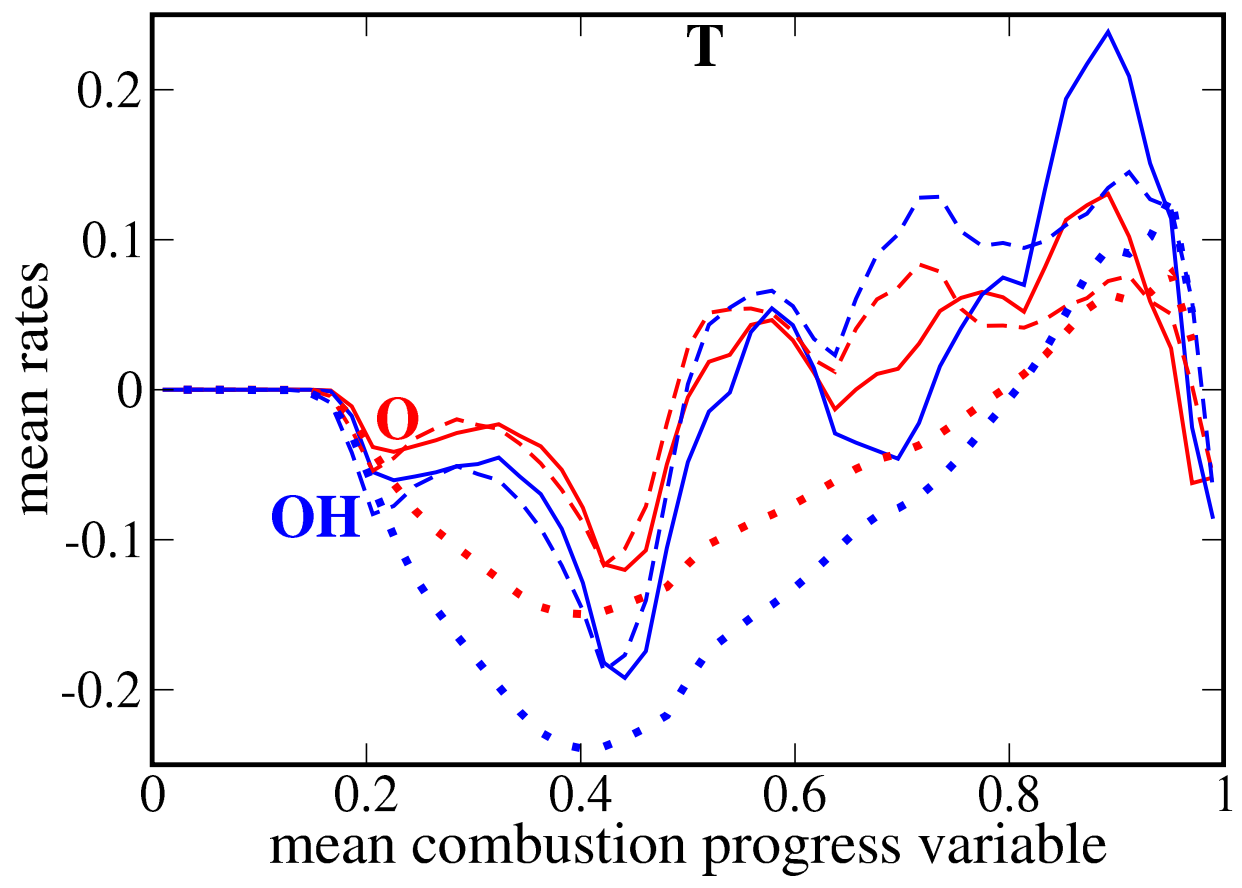
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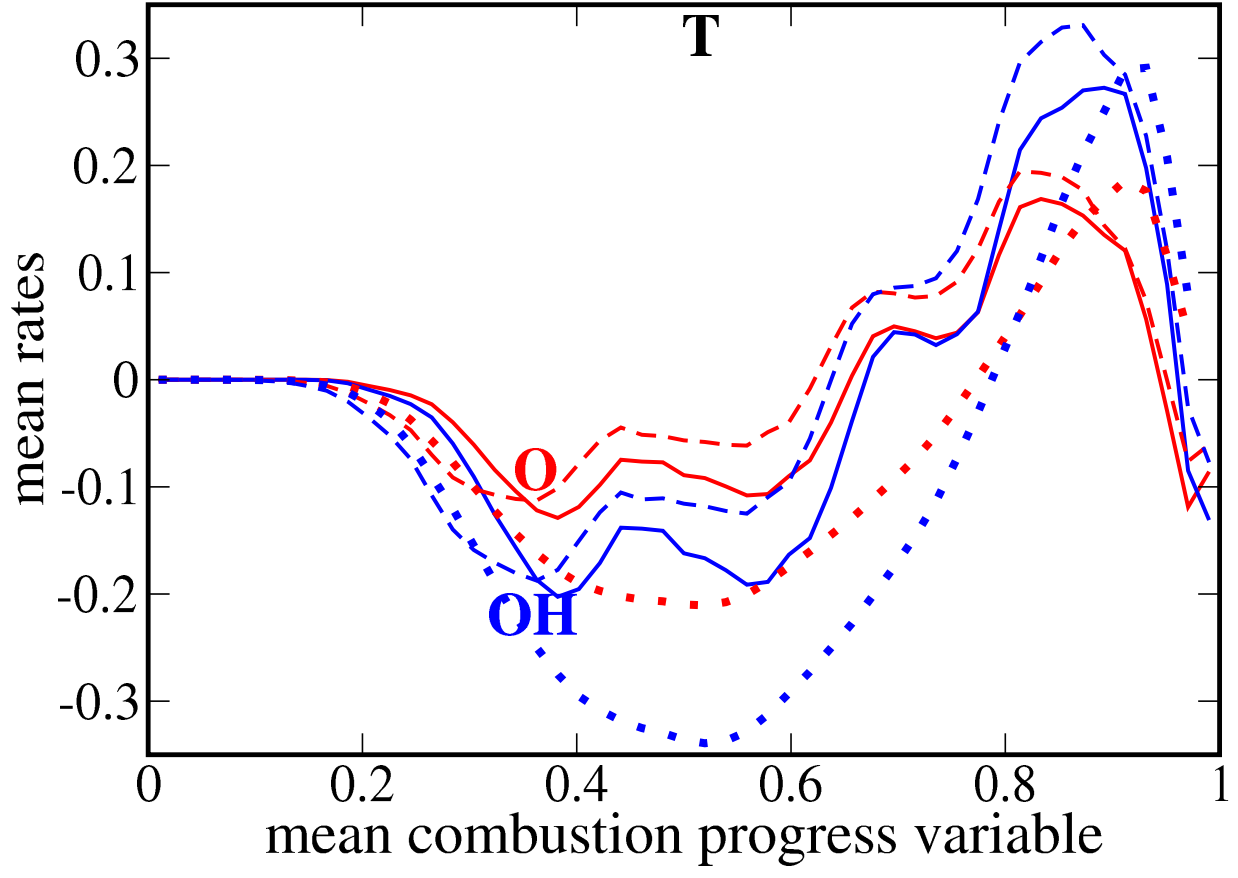


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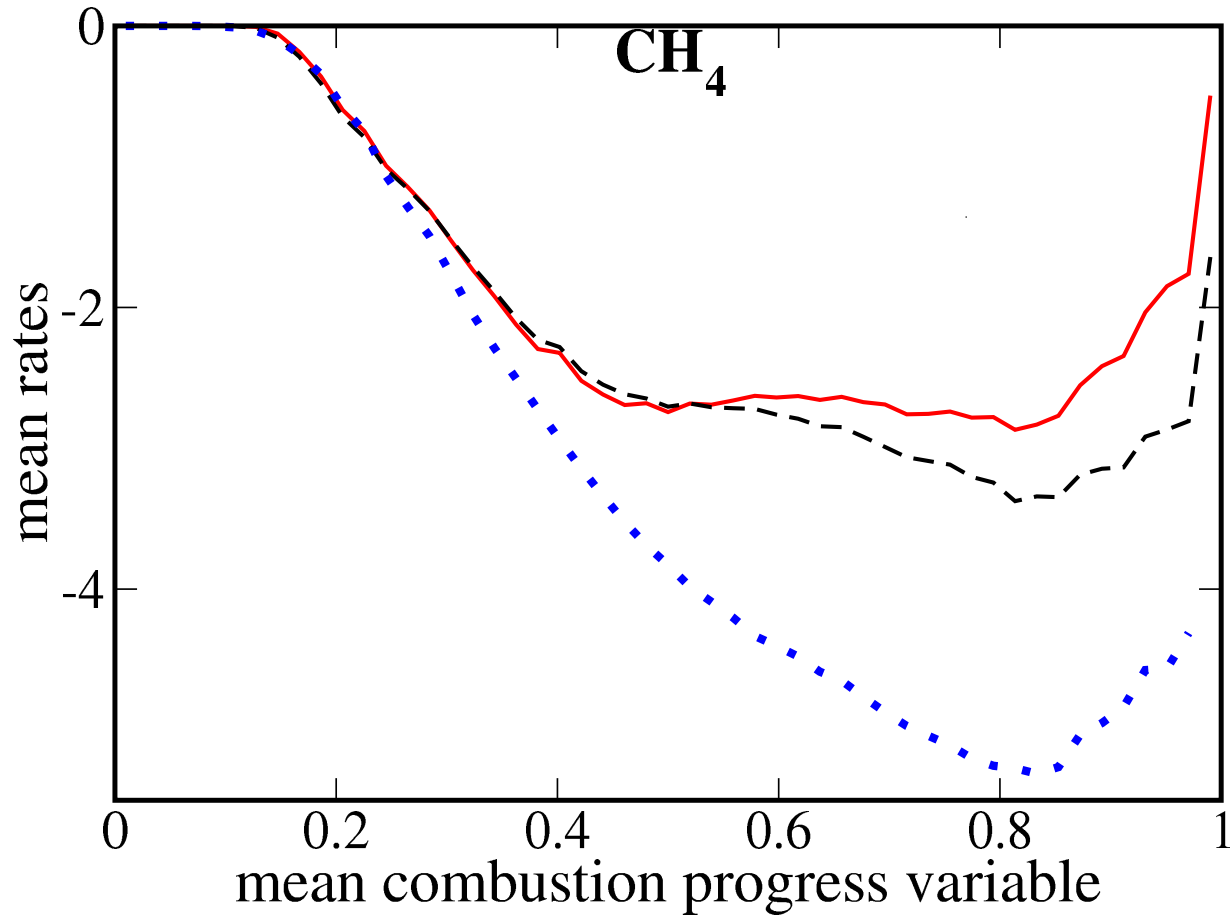
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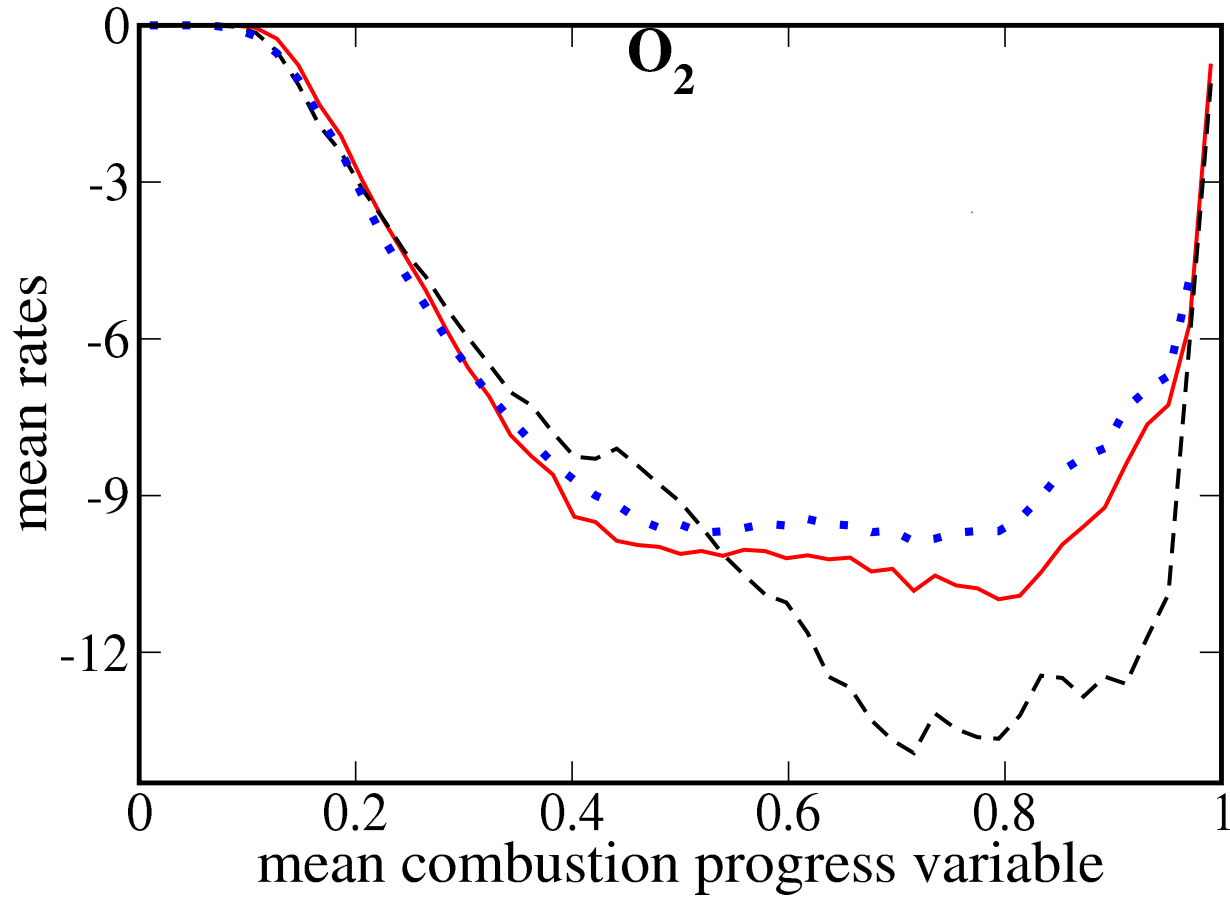
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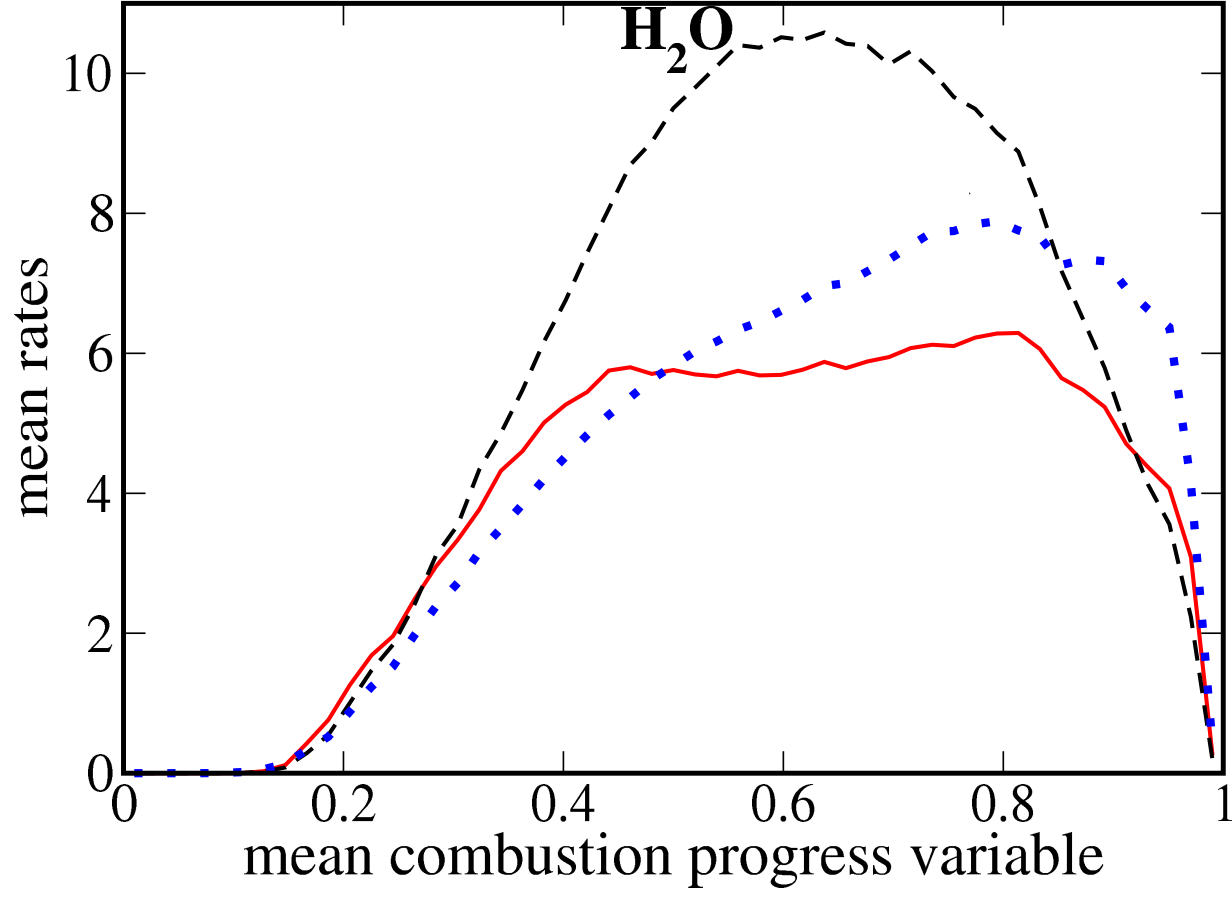
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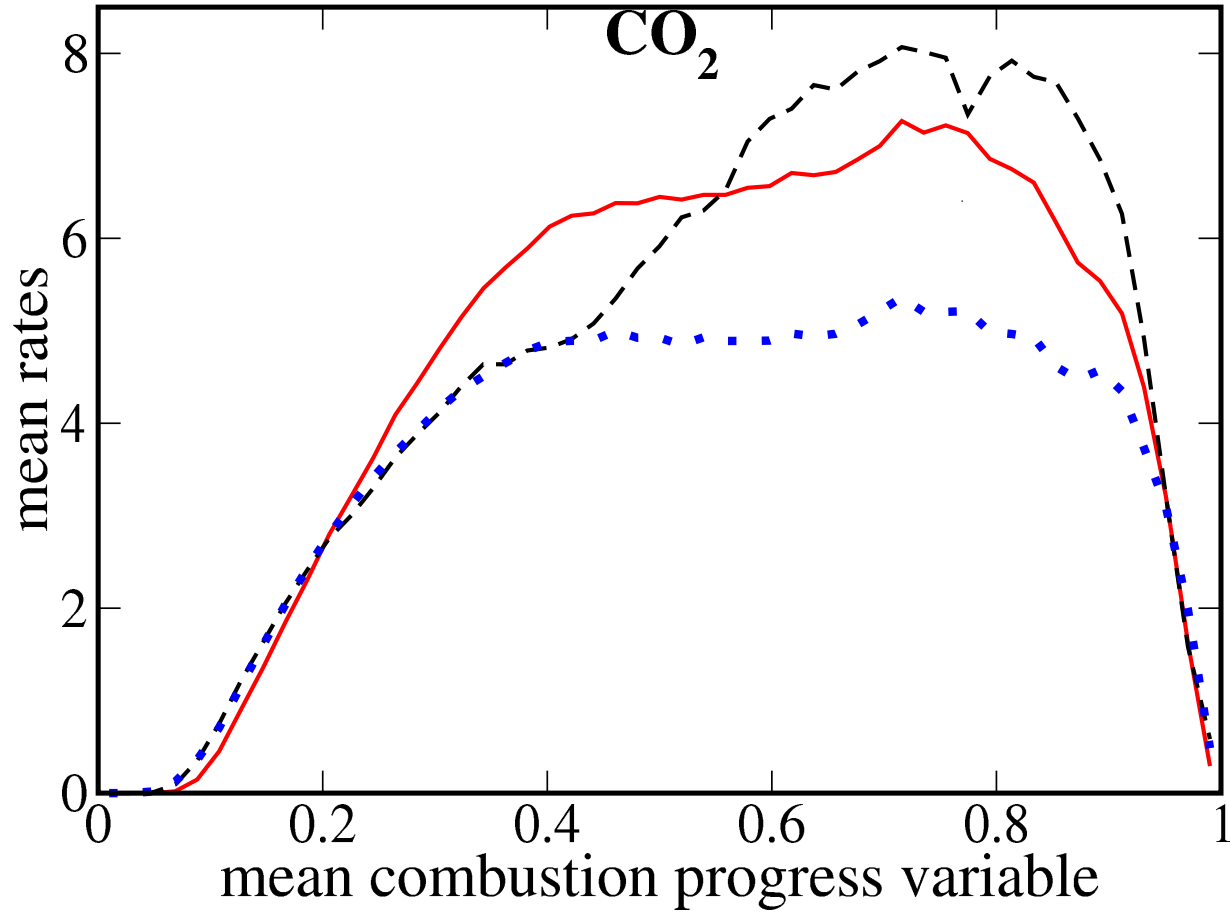
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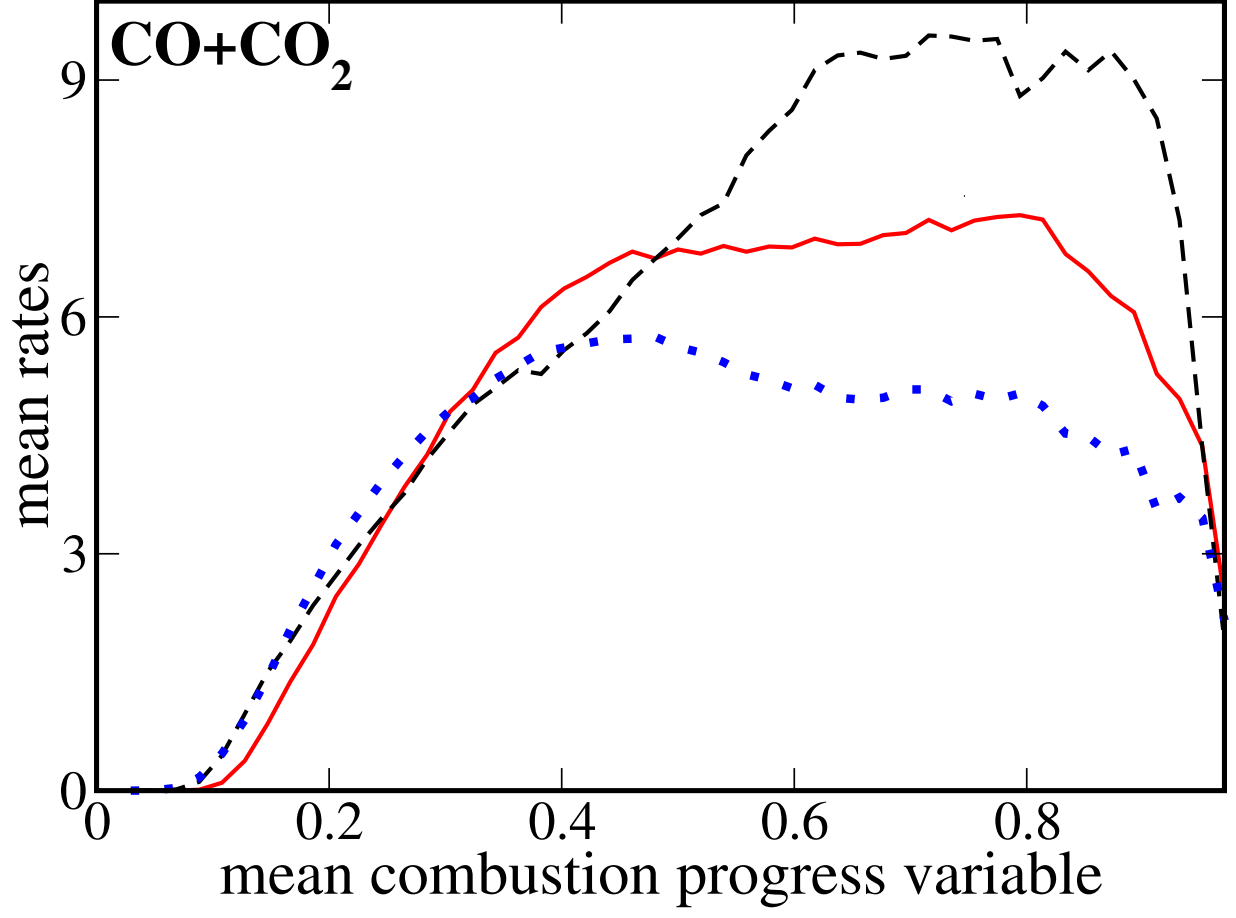
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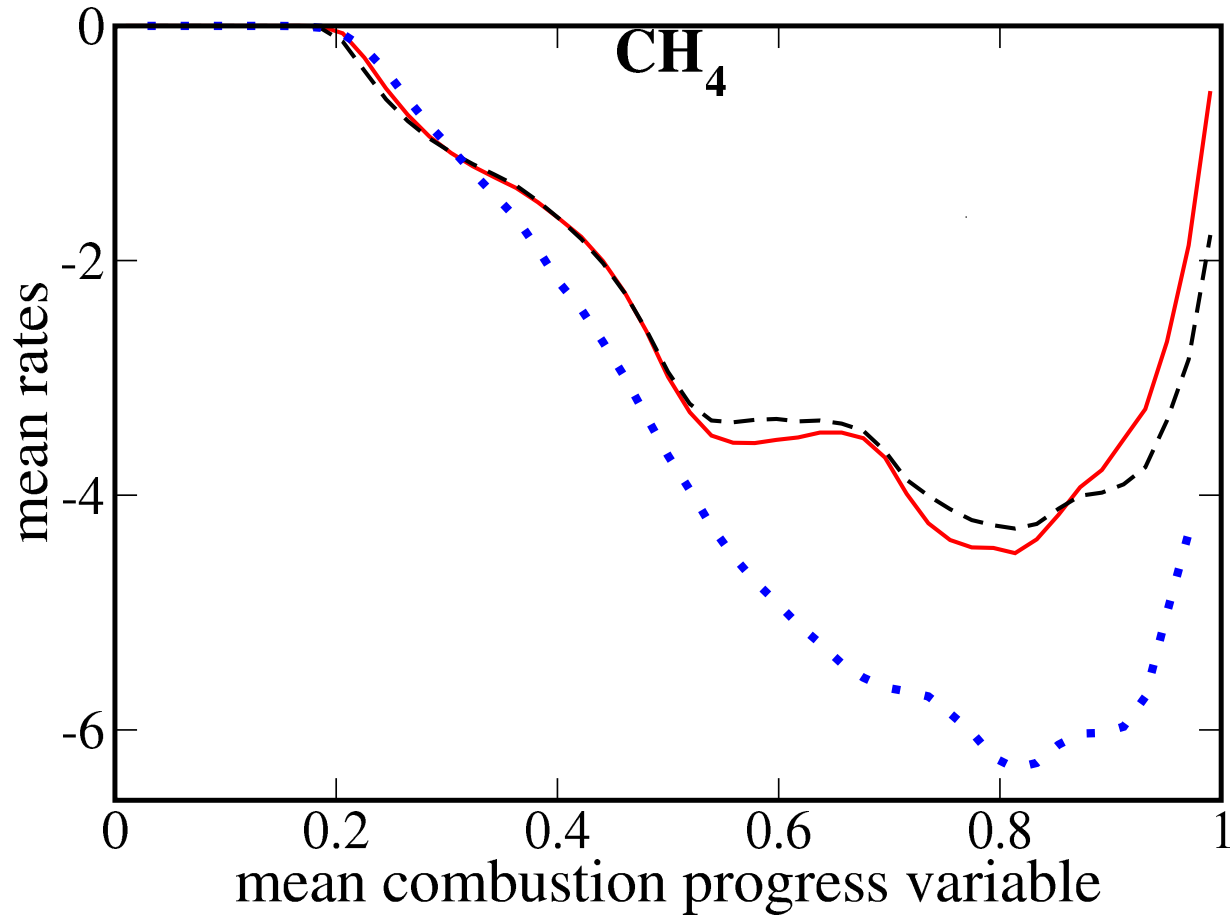
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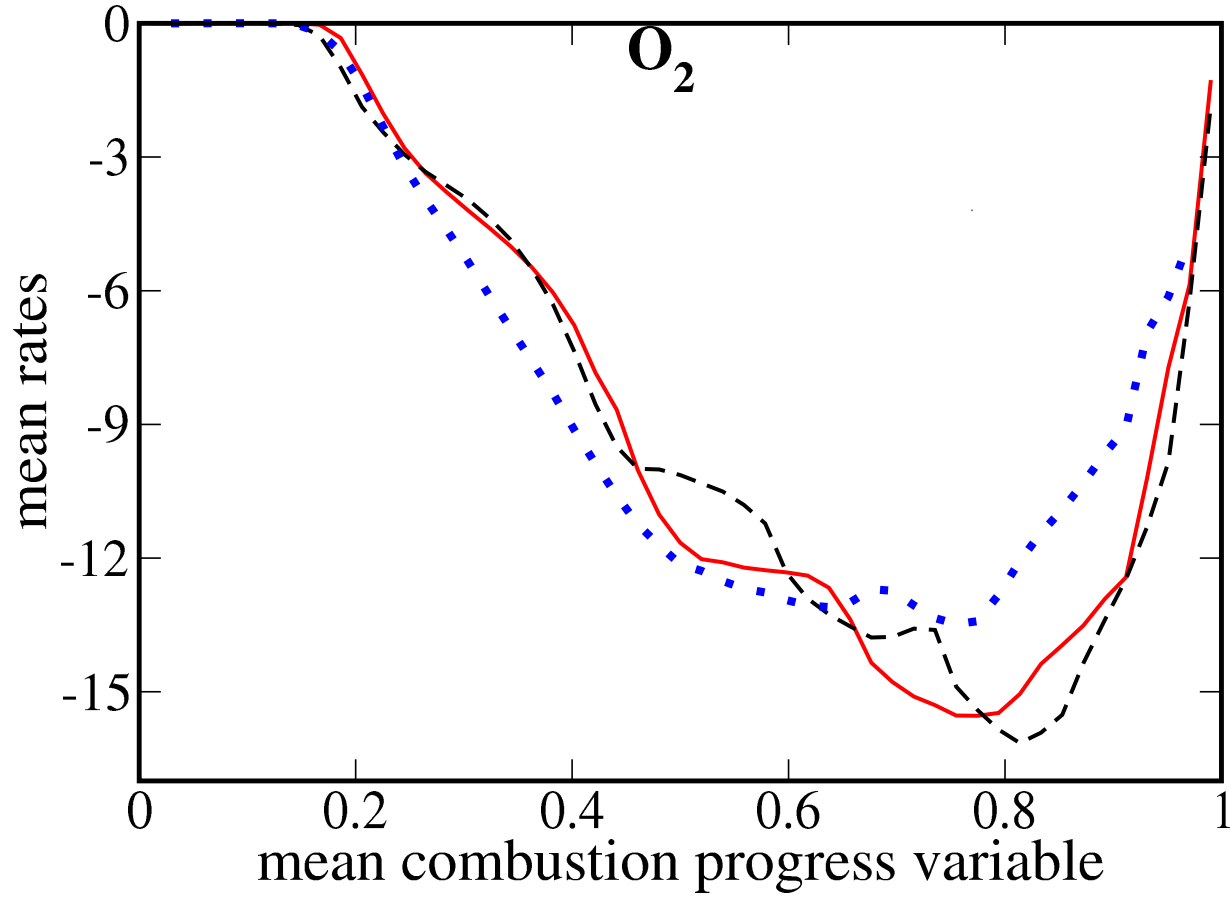
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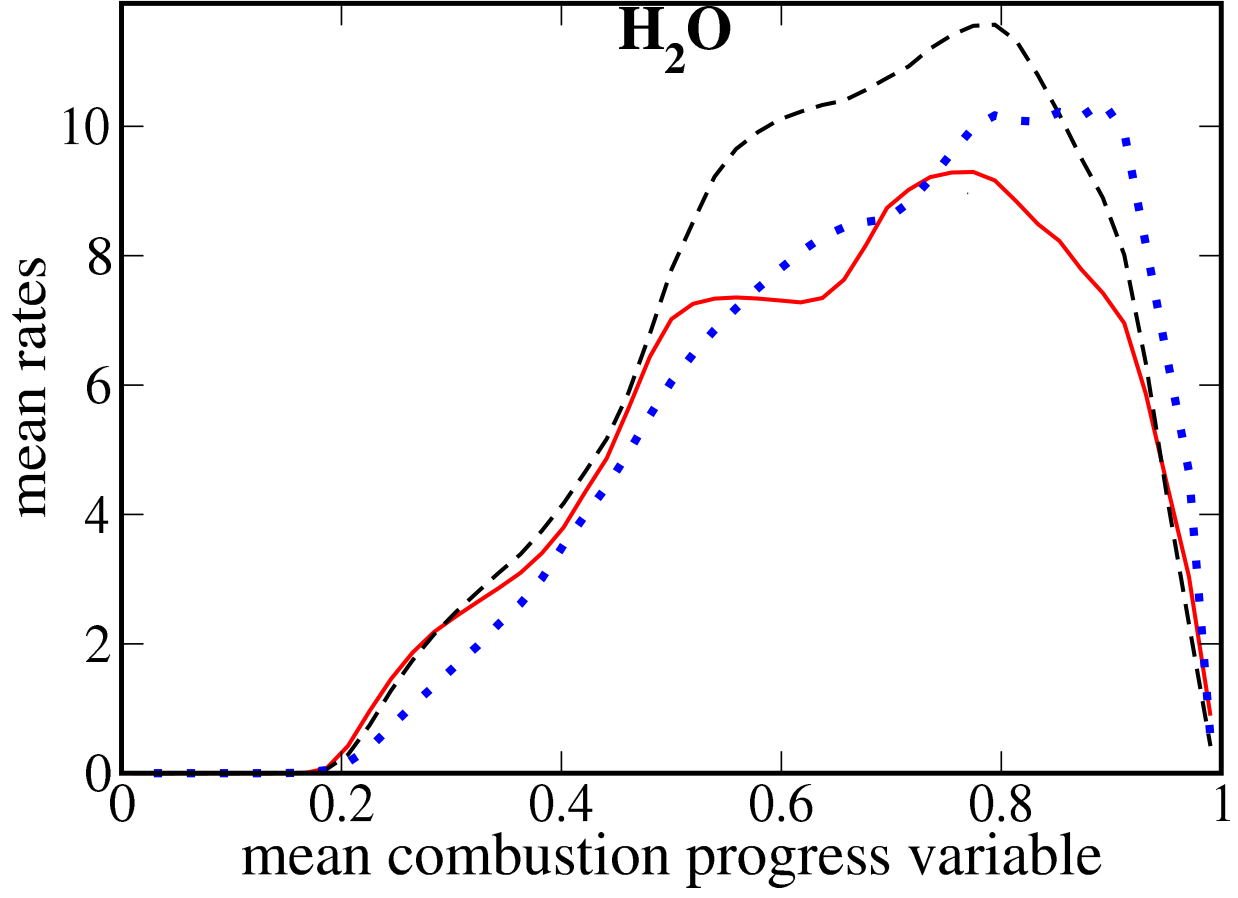
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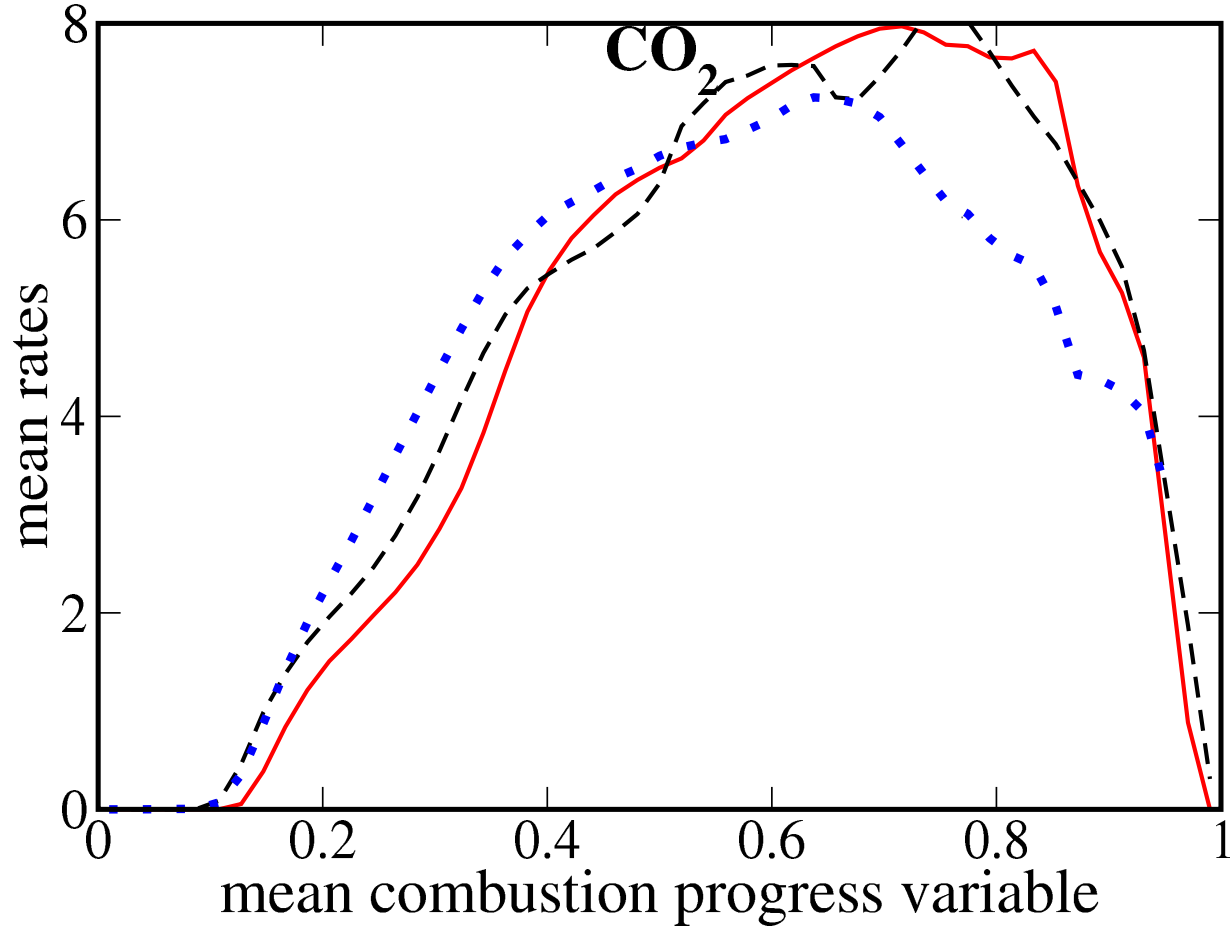
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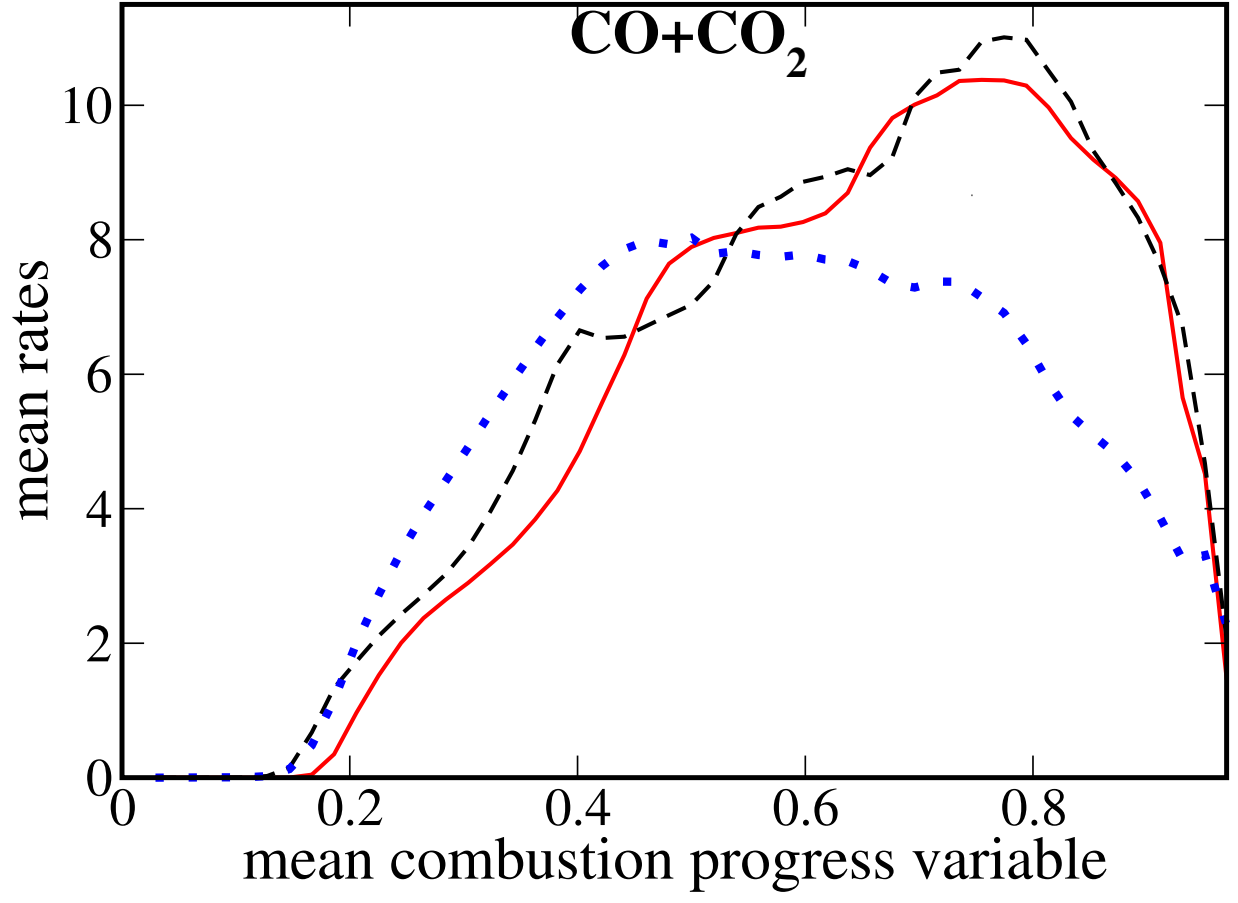
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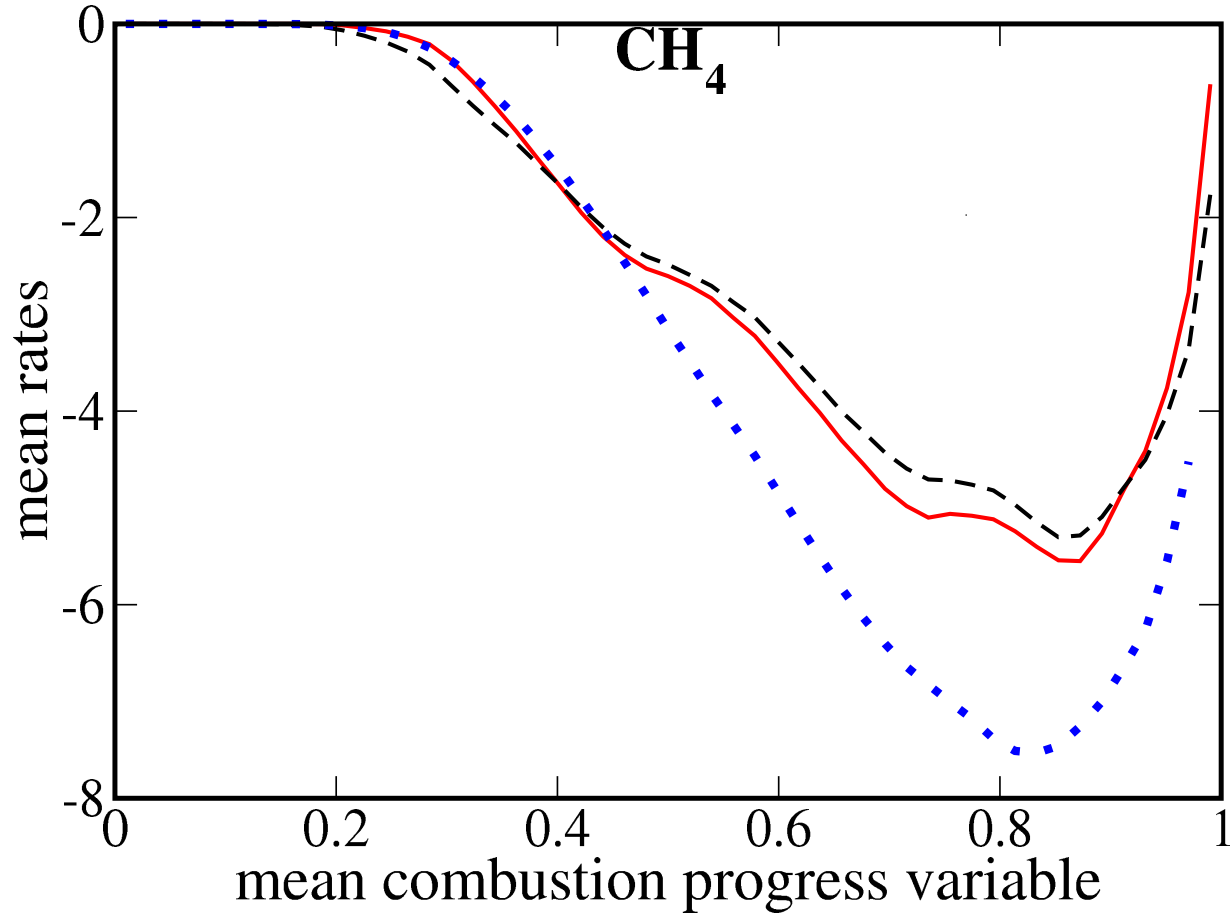
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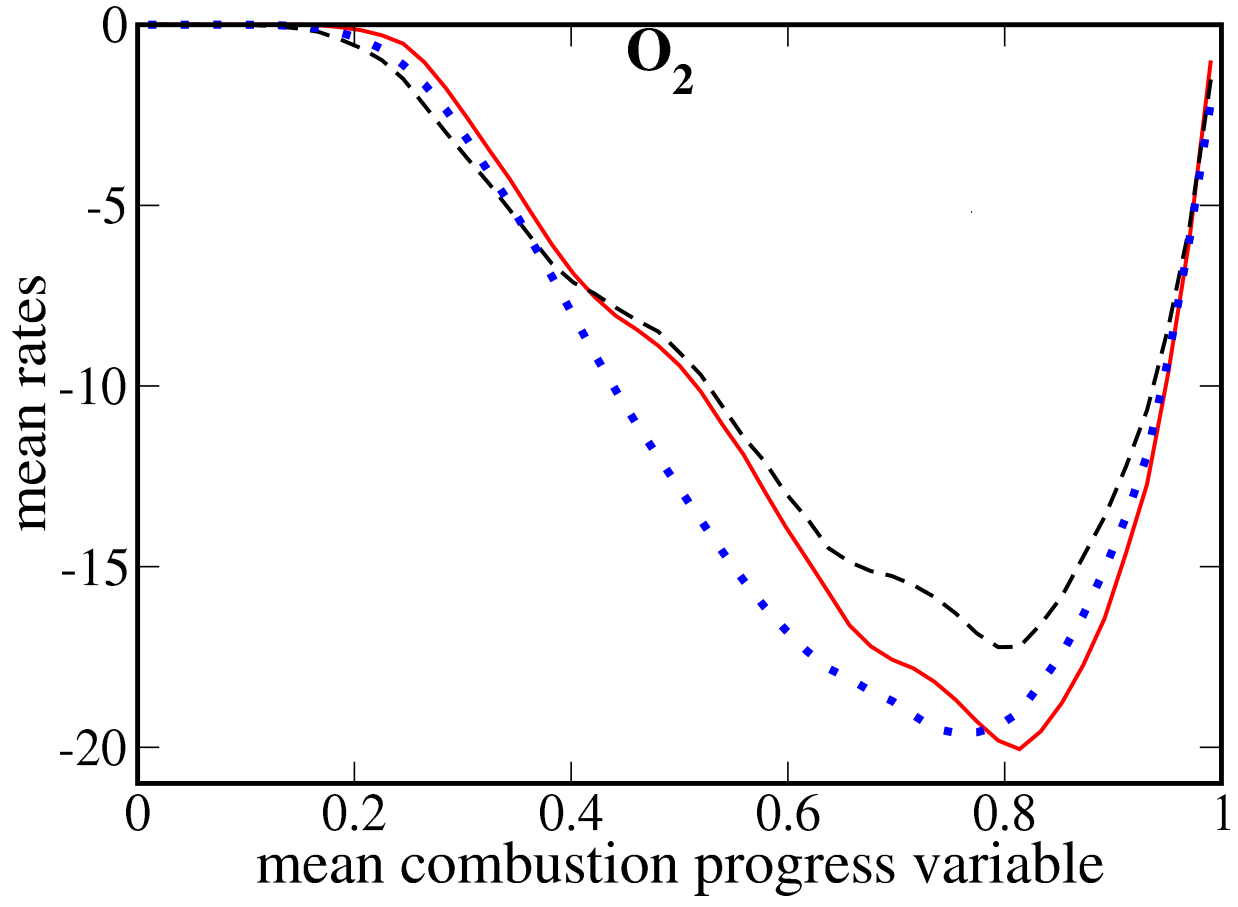
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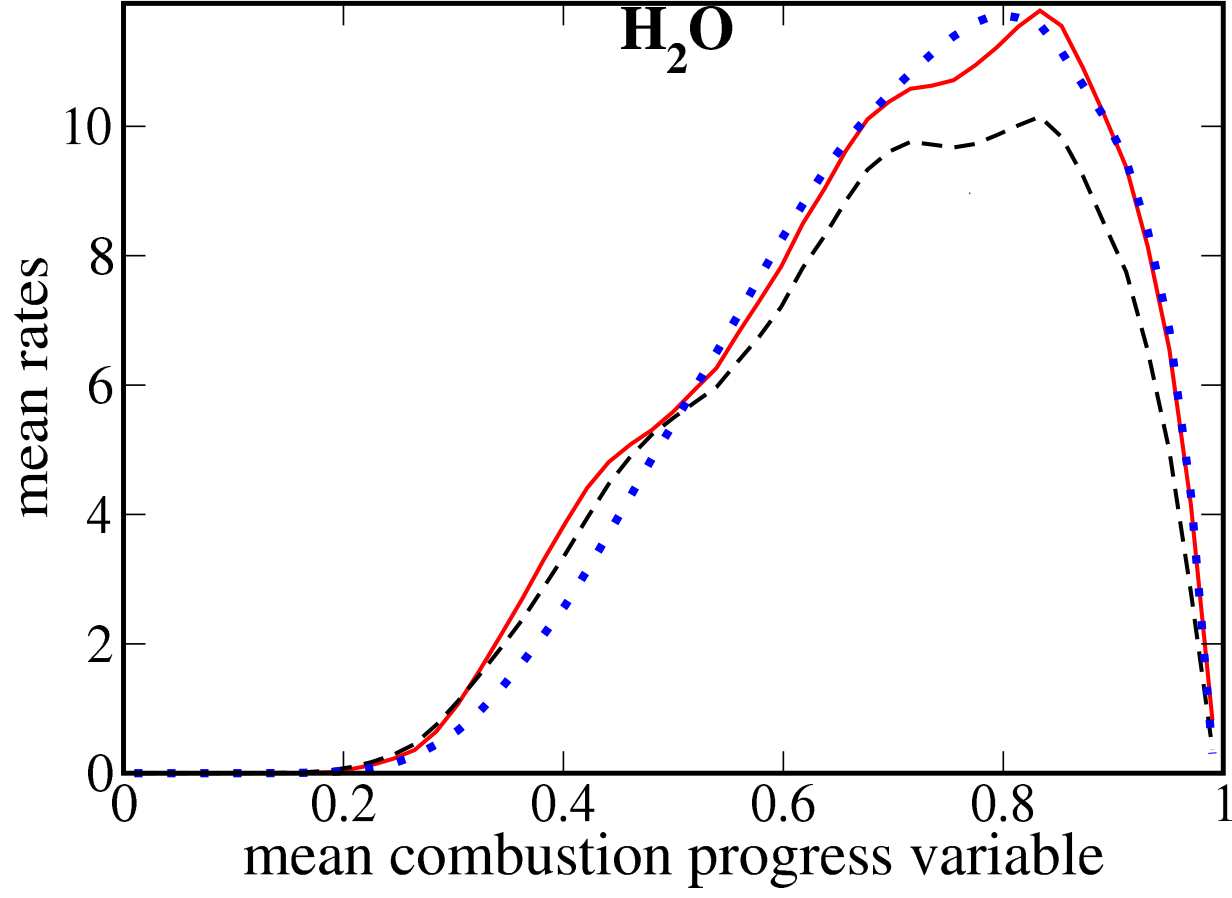
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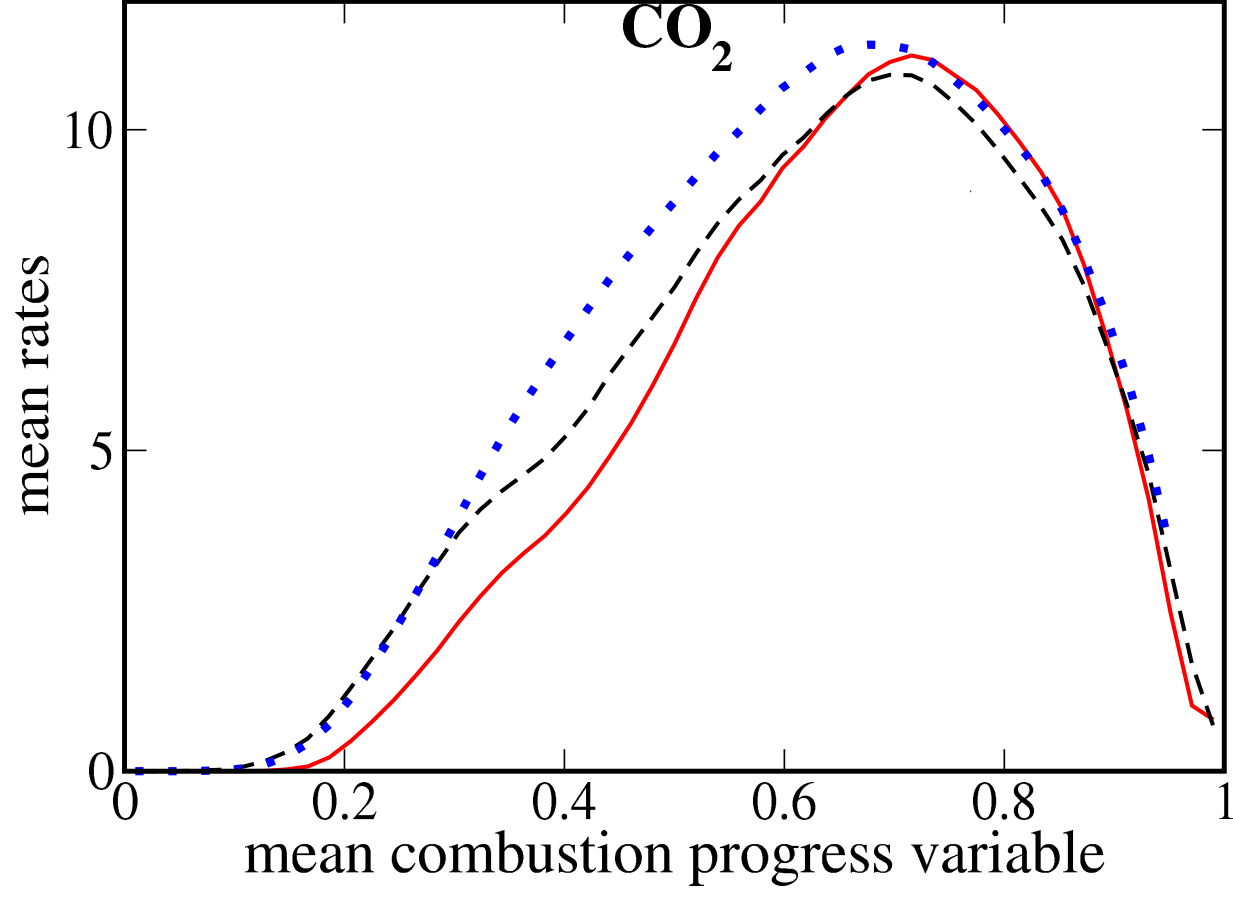
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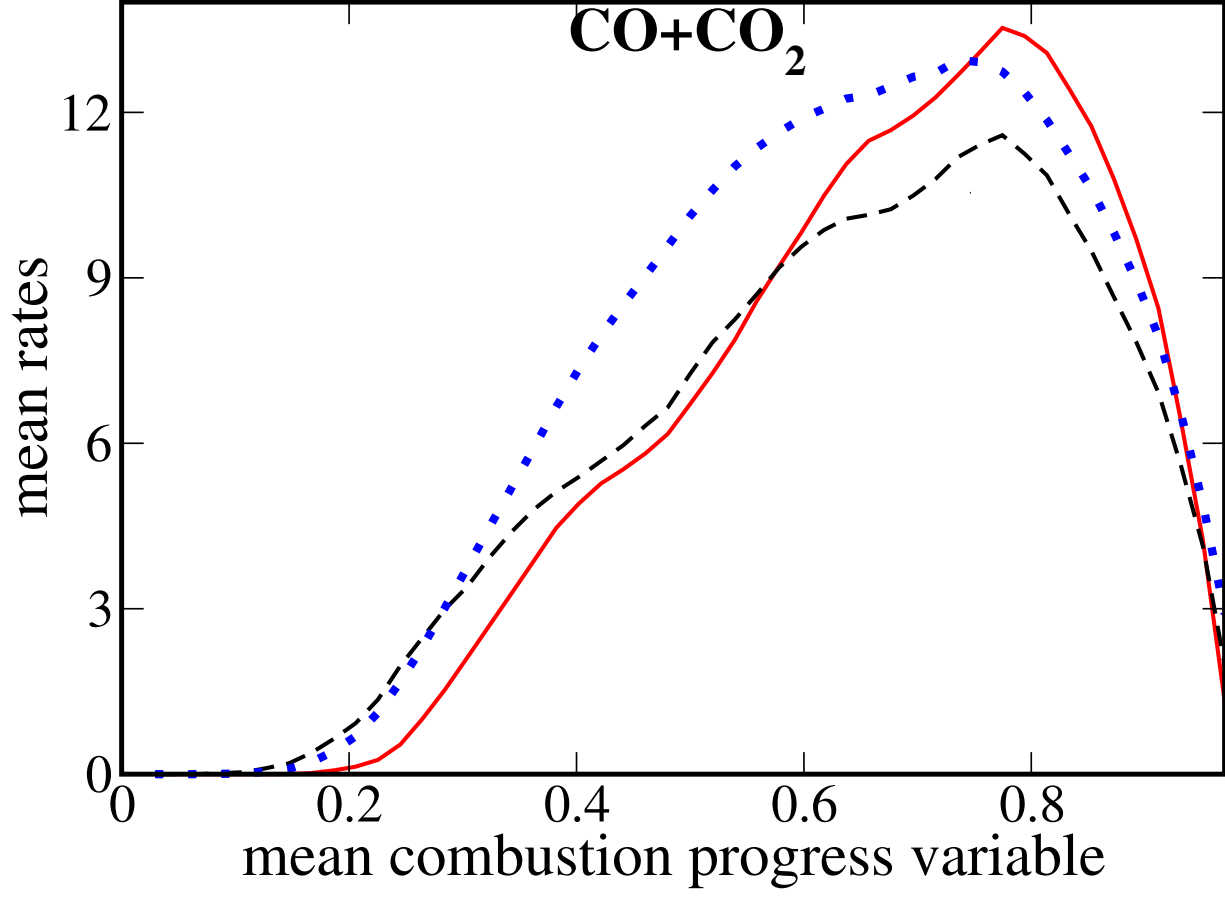
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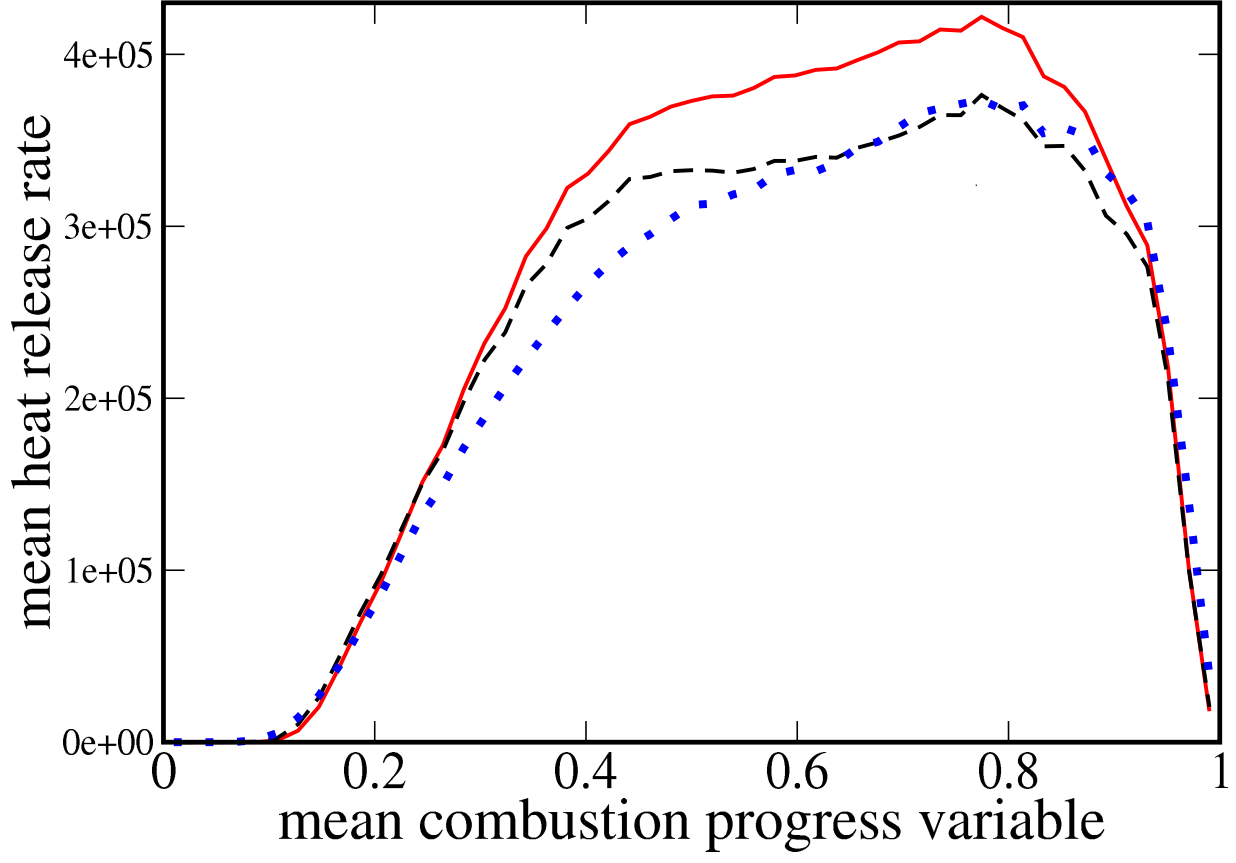
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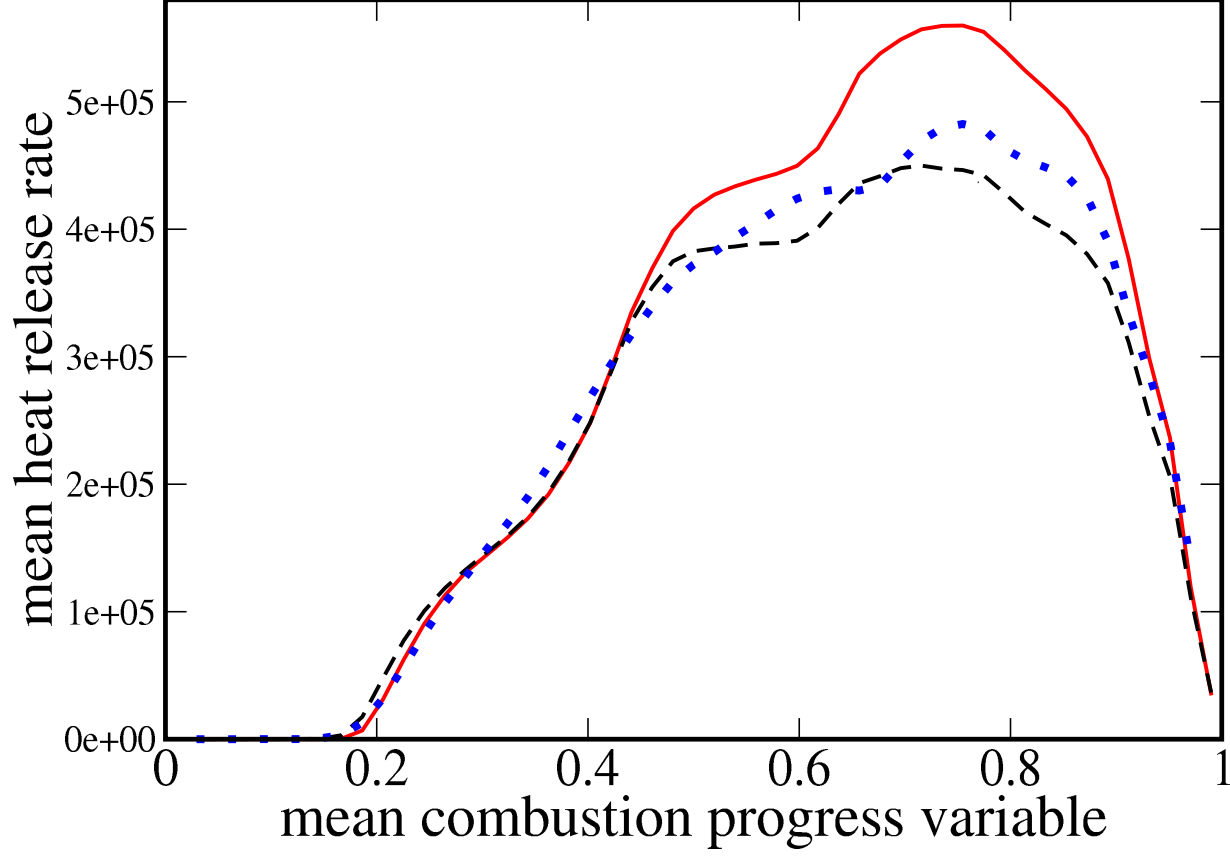
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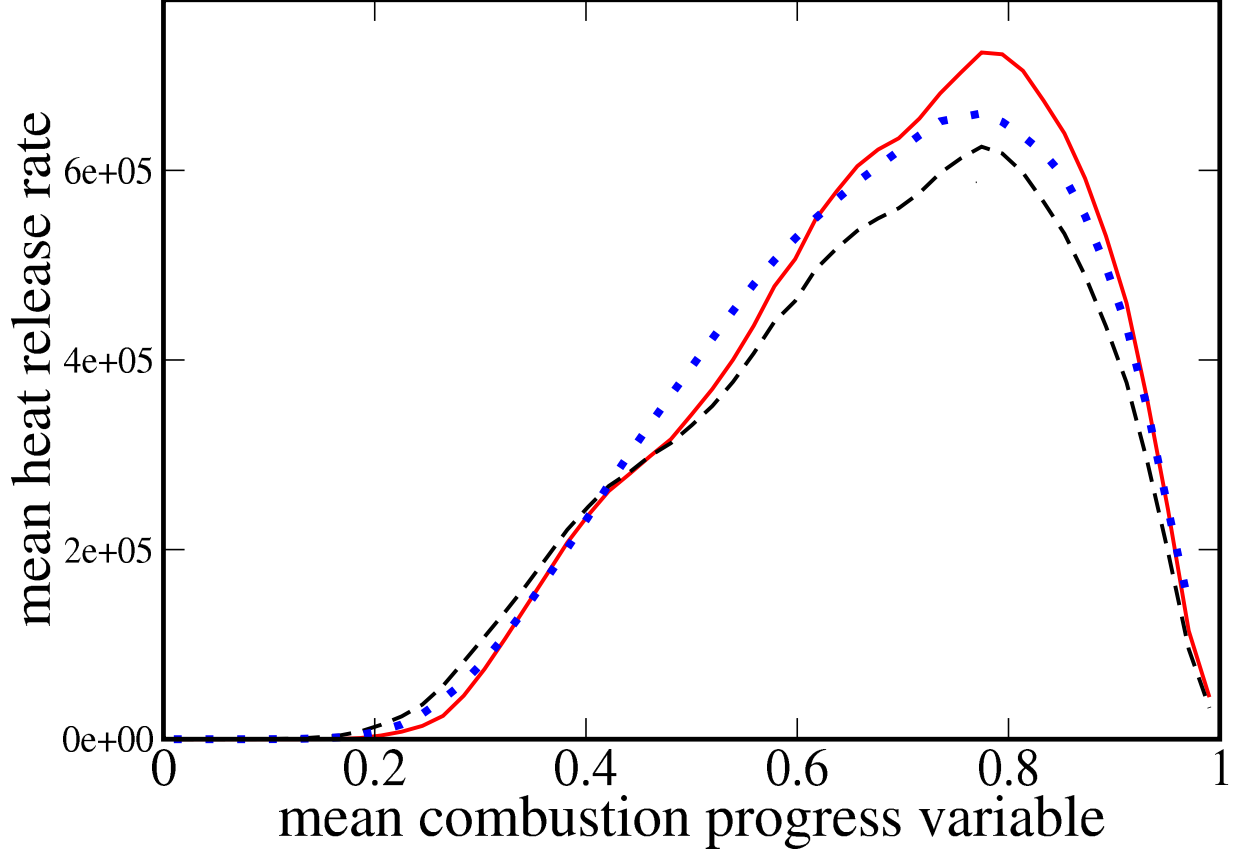
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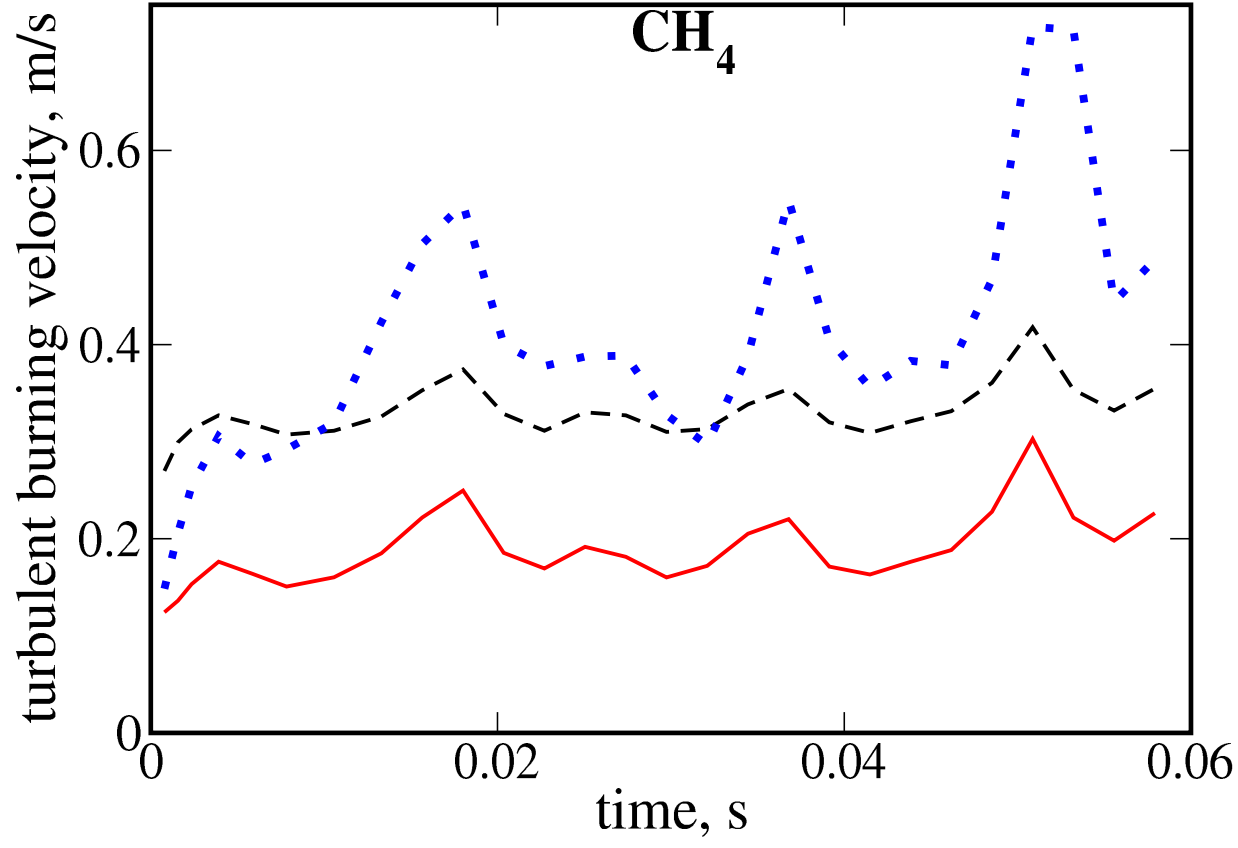
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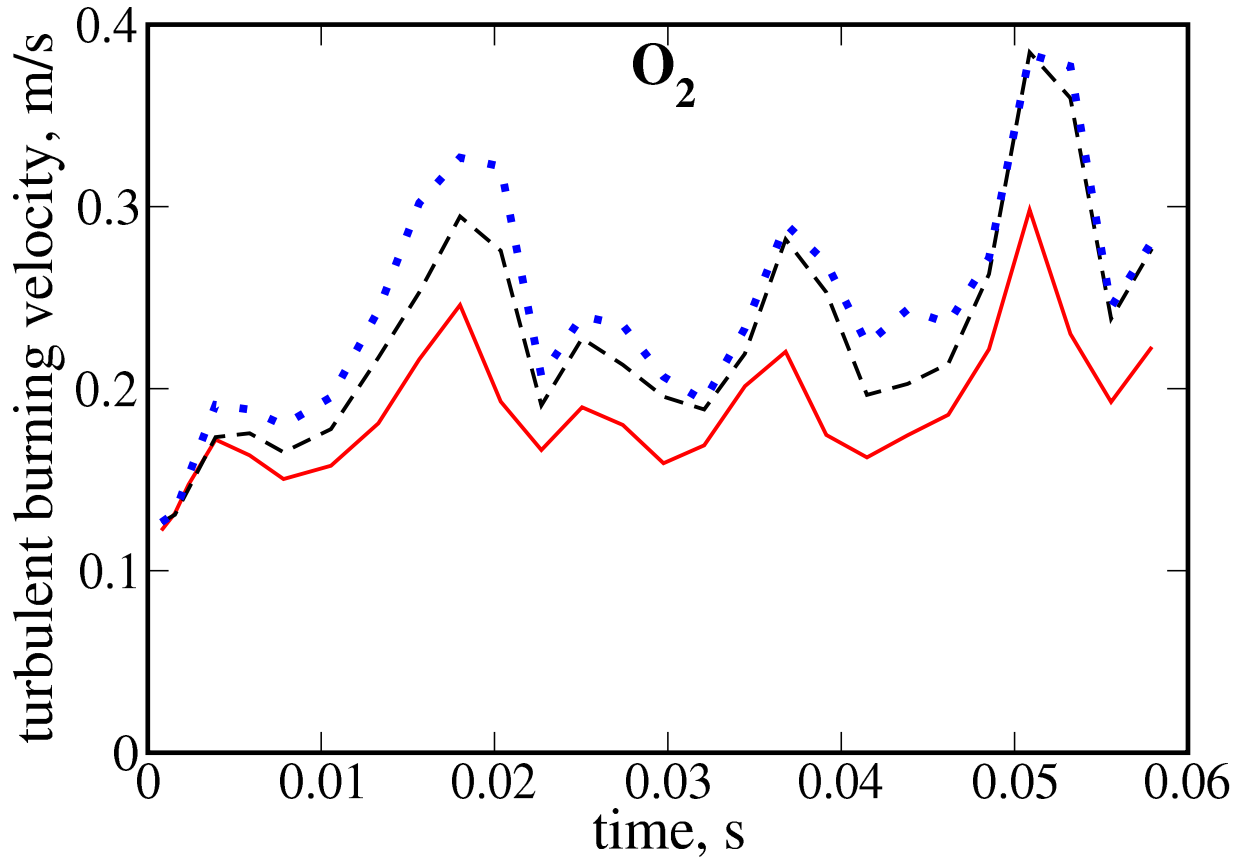
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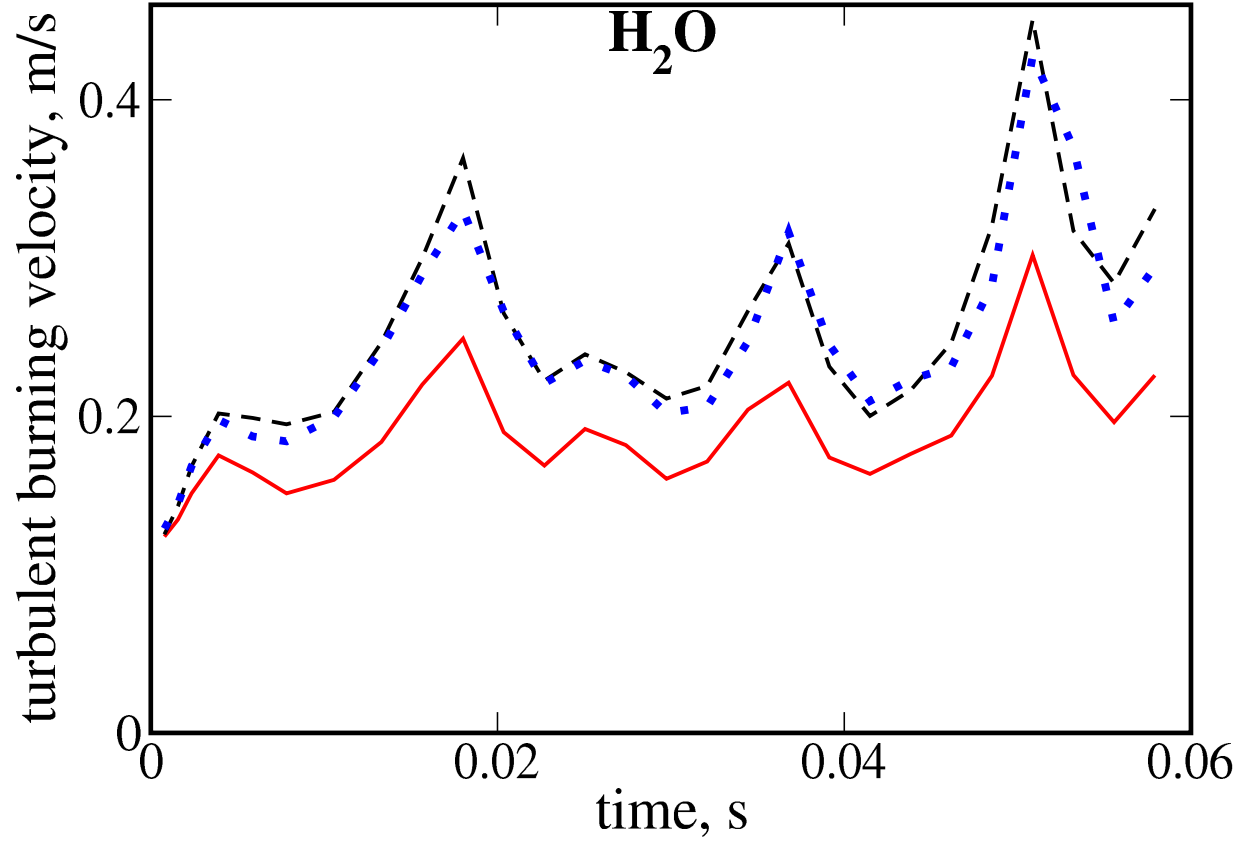
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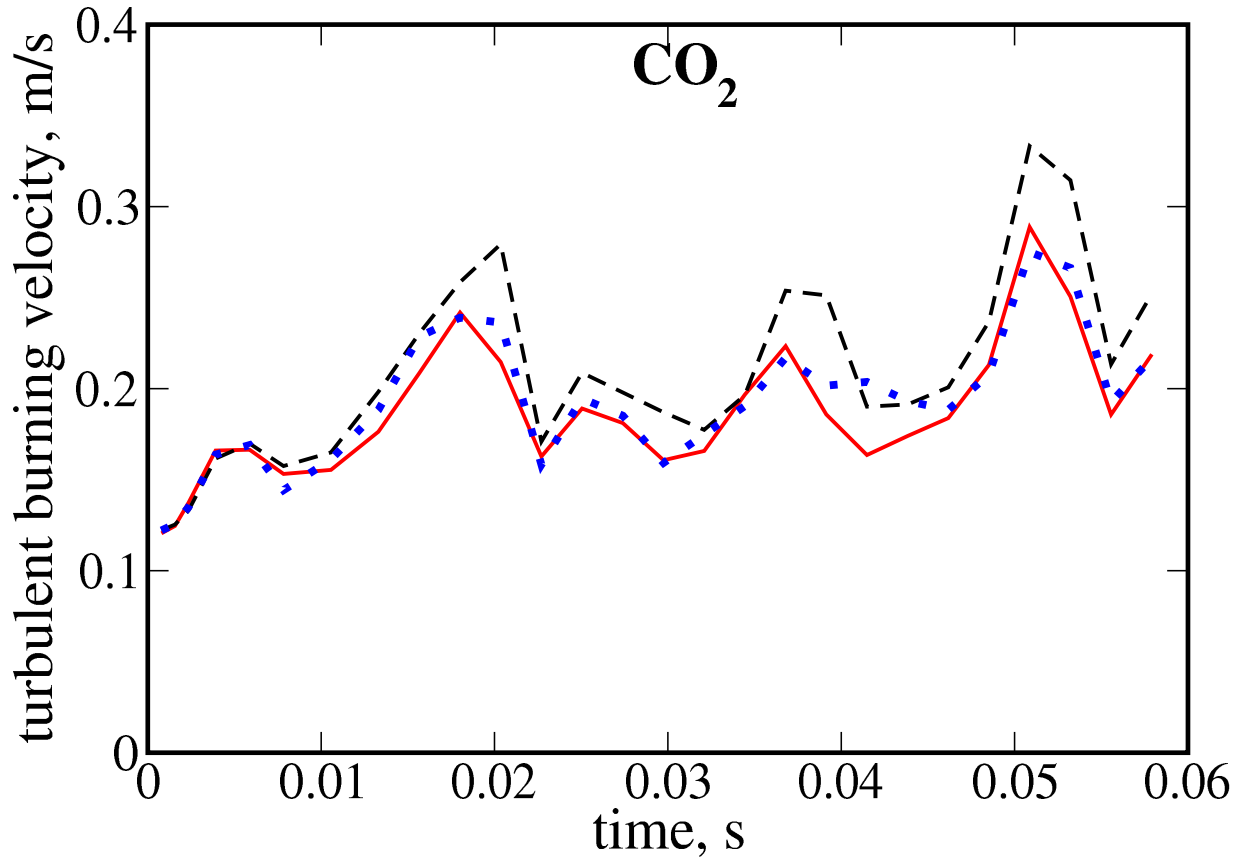
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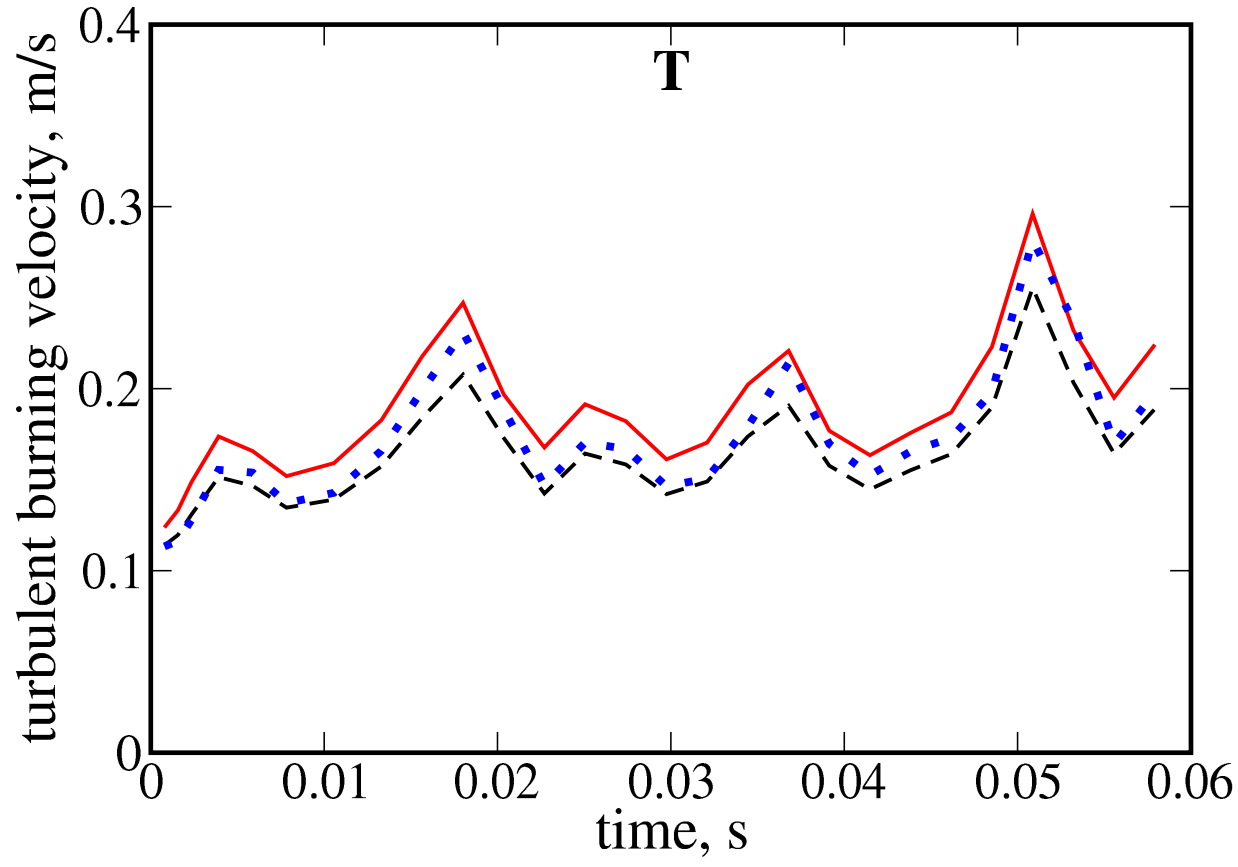
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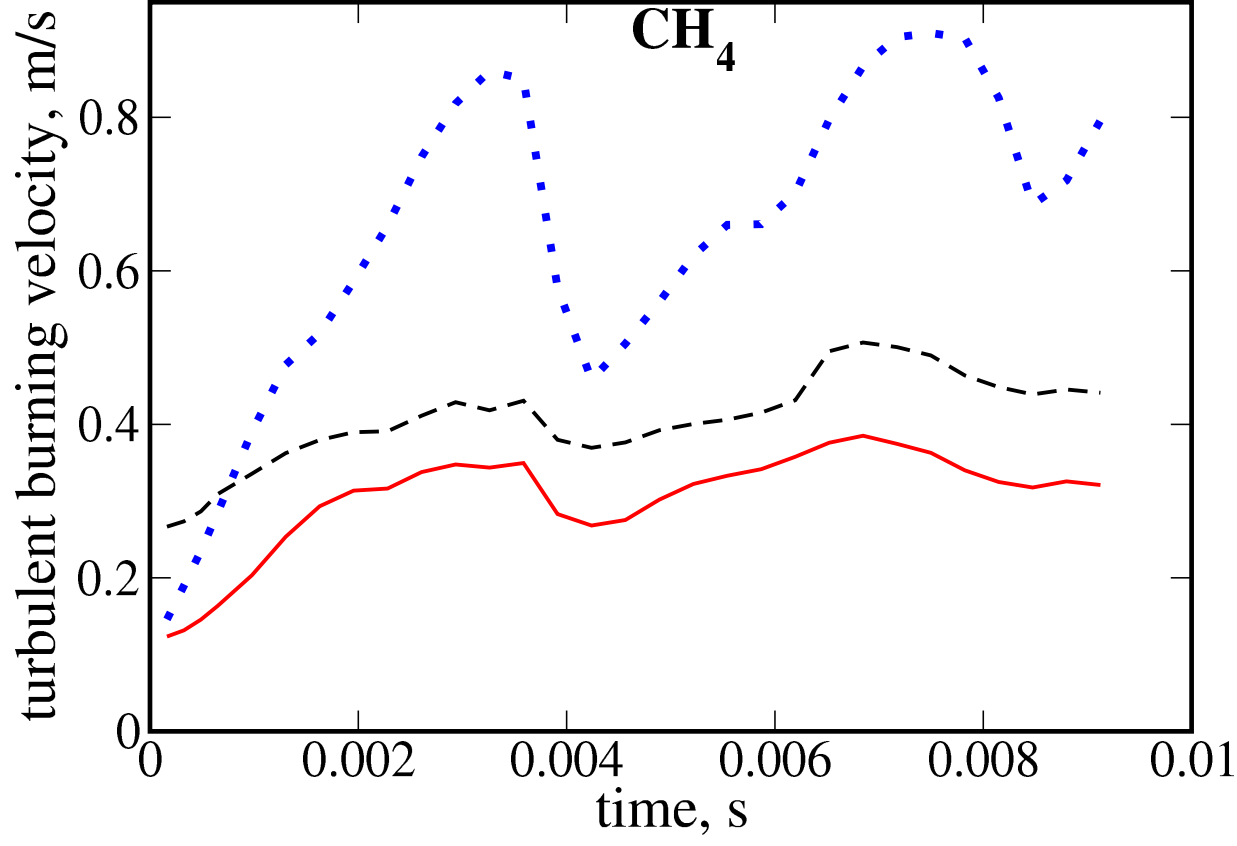
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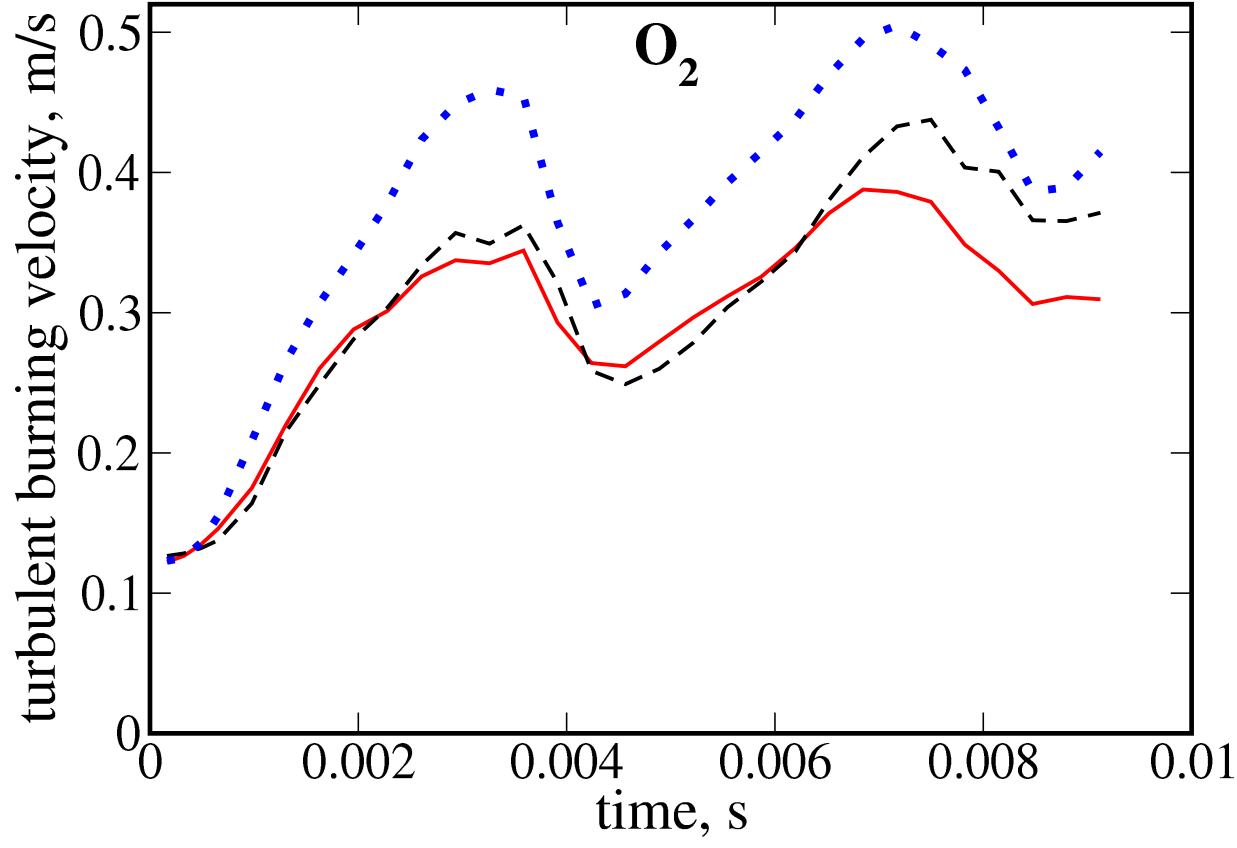
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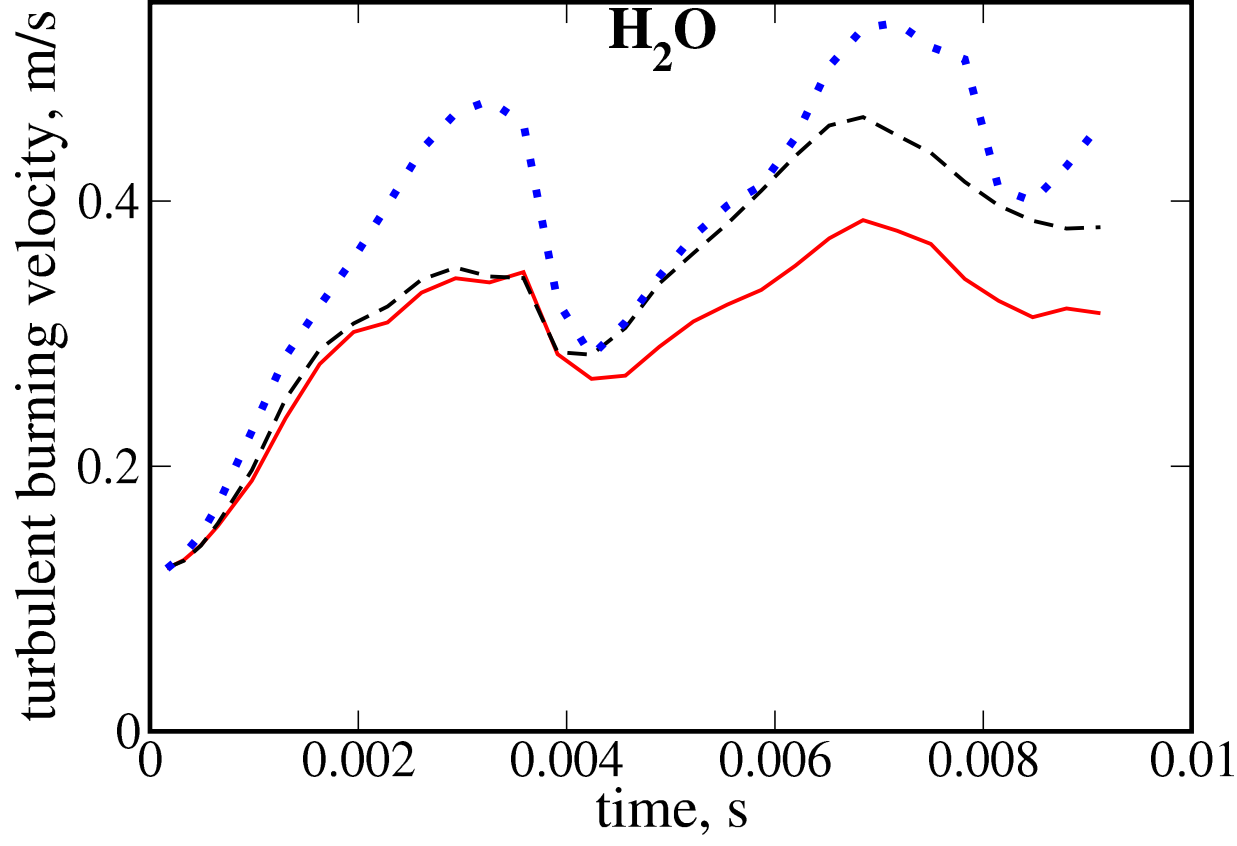
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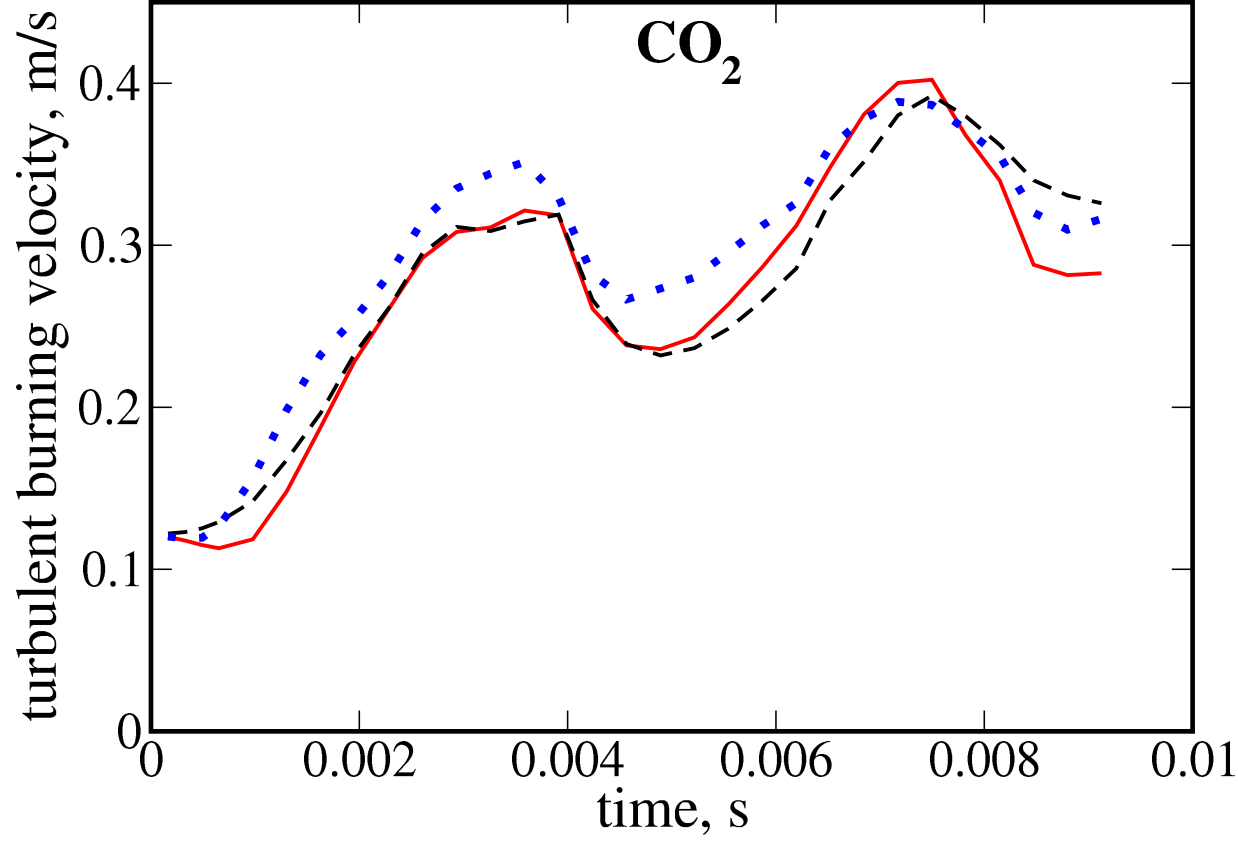
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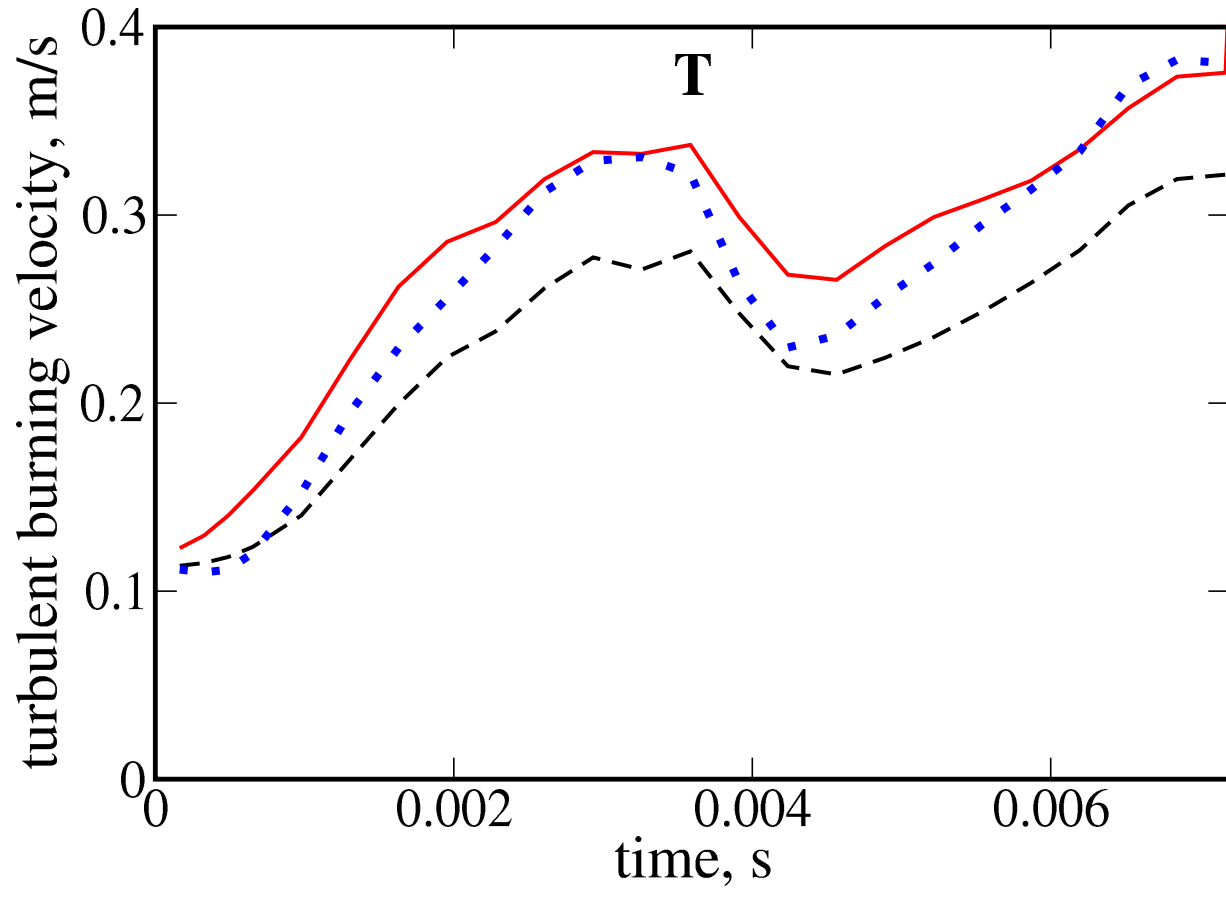
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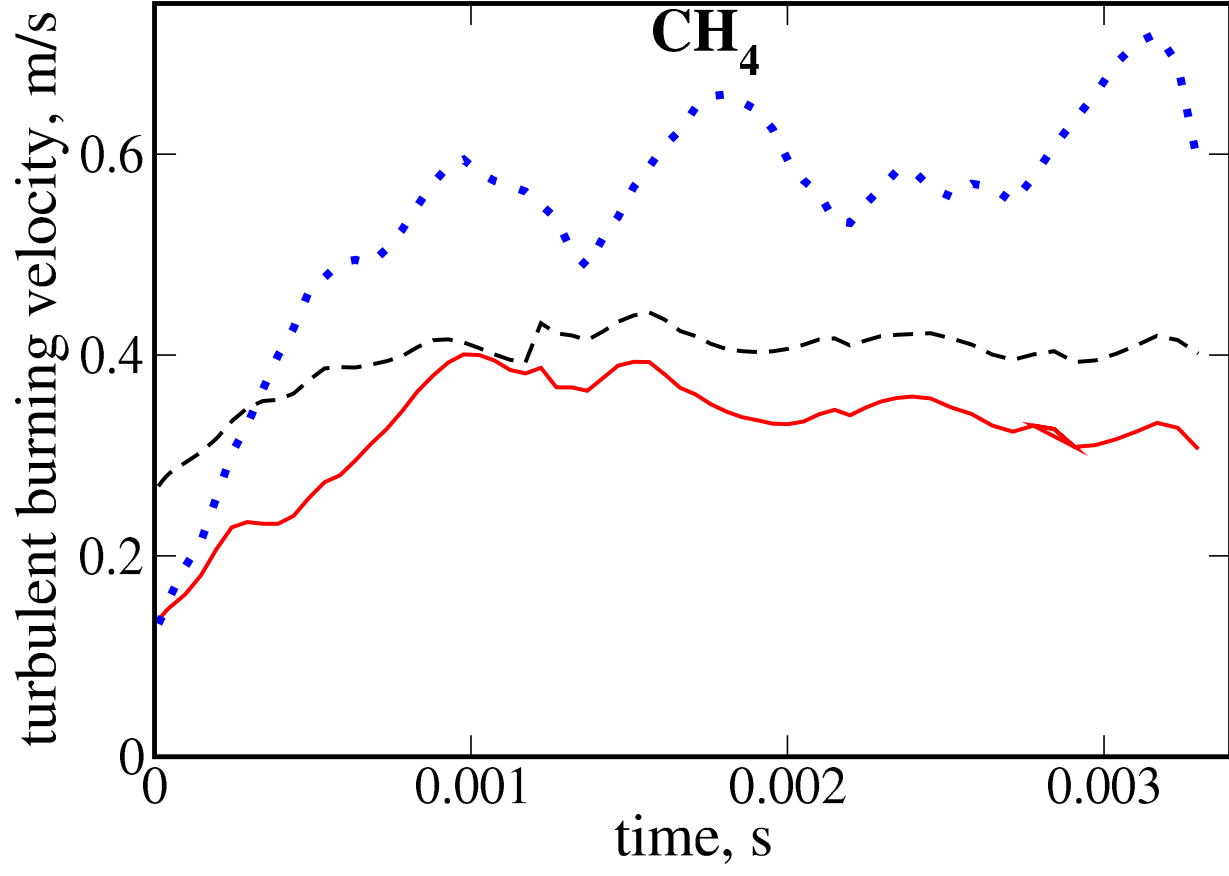
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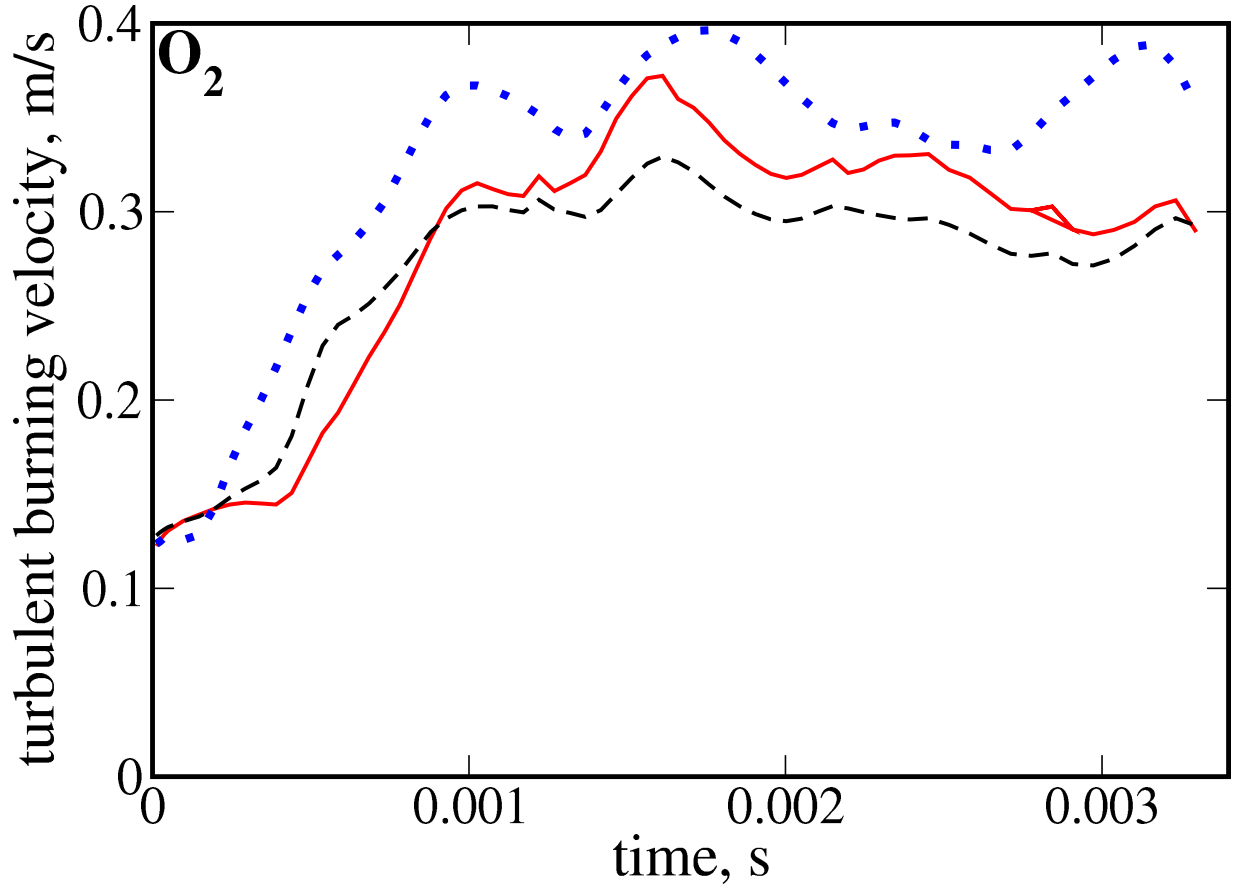
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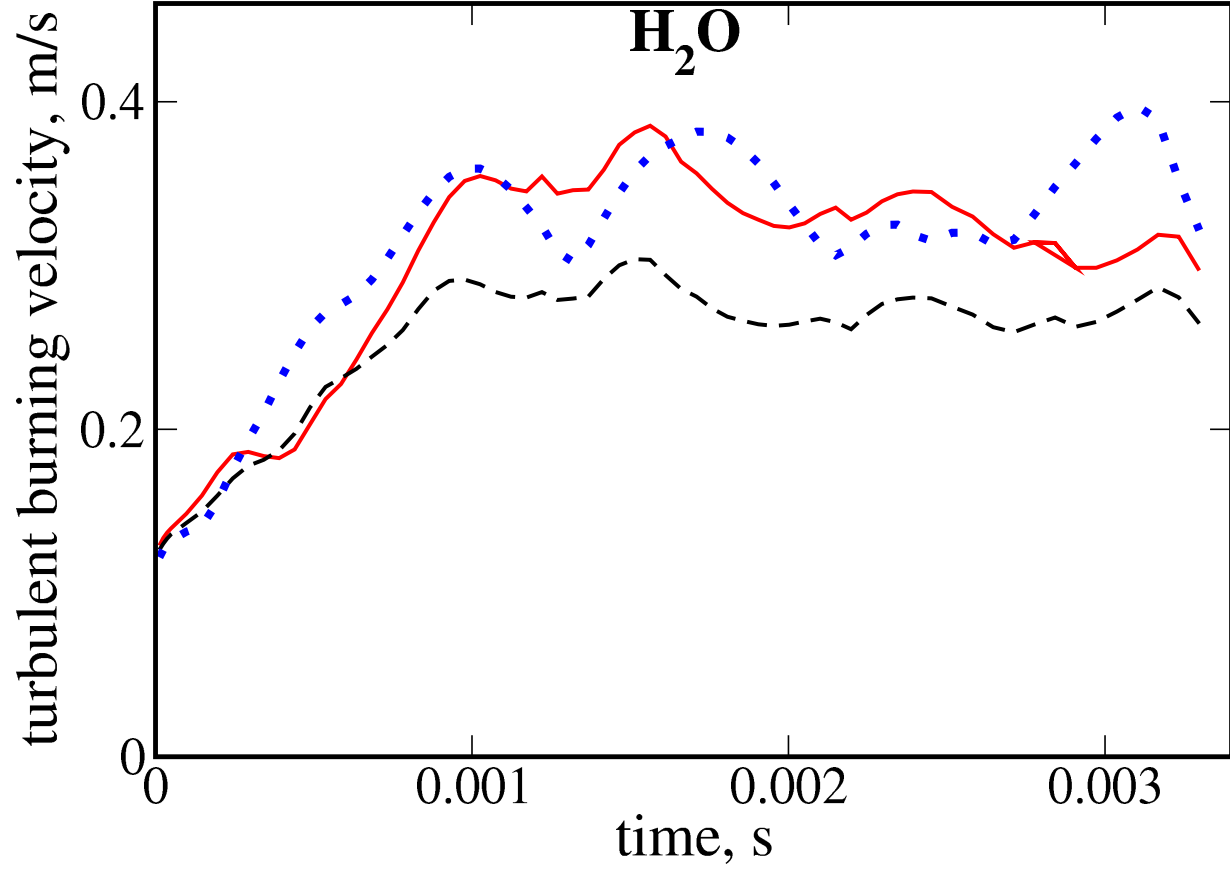
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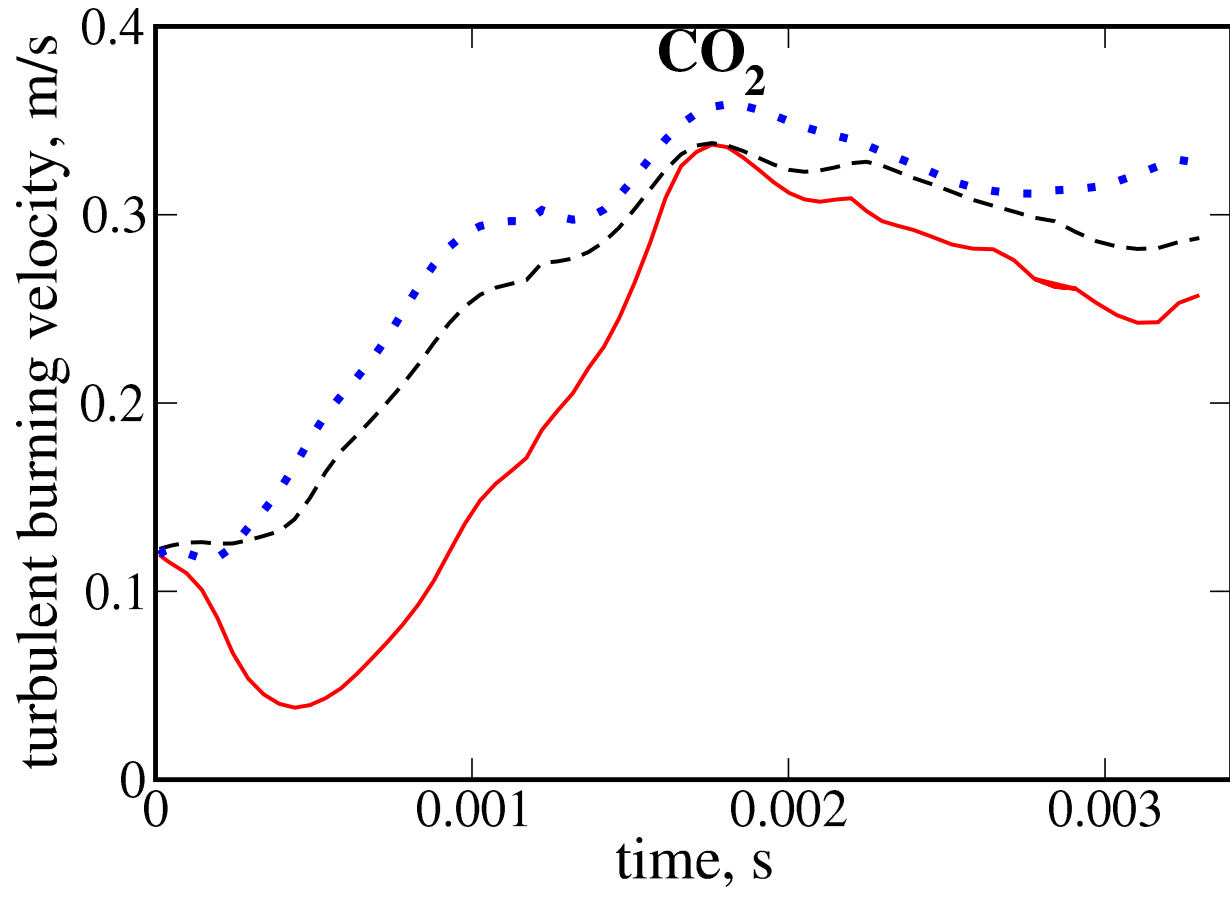
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